INFLUENCE OF NITROGEN FERTILIZER AND STRAW RETURNING ON CH_4 EMISSION FROM A PADDY FIELD IN CHAO LAKE BASIN, CHINA

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Abstract. In the rice paddy ecosystem, application of nitrogen fertilizer and straw to soil methanogenic bacteria provides abundant methanogenic substrates, which significantly influences methane (CH_4) emission from paddy fields. The effects of nitrogen fertilizer and straw returning on CH_4 emissions in rice paddy fields were studied during a two year of the field experiment in Chao lake basin, China. The experiment consisted of 4 treatments: Control (CK), Traditional fertilizer (CT), Optimized fertilizer (CO) and CO with straw-return (CO + SR). The cumulative effects of straw-returning practices on greenhouse gas emission in a rice-wheat rotation system were determined, along with an estimation of CH_4 in a rice growing season. According to our results, The CH_4 emission fluxes from paddy field showed three different peak trends; Compared to CK, CT, CO, and CO + SR increased seasonal CH_4 emission by 36.6%, 45.8% and 42.0% in 2013 and by 42.0%, 48.5% and 80.1% in 2014, respectively. Anaerobic decomposition of wheat straw accelerates the decline of soil redox potential (Eh) after flooding, thereby providing suitable environmental conditions for the growth of methanogens and promoting CH_4 production in the subsequent rice season. The CH_4 emission fluxes of CK, CT, CO, and CO + SR were significantly correlated with soil temperatures at 5 cm depth. Therefore, our findings show that application rates of nitrogen fertilizer and straw returning to a paddy field could significantly affect the CH_4 emissions in China.

Keywords: *CH_4 flux, rice paddy, chemical fertilizer, straw application, nitrogen use efficiency*

Introduction

Since the industrial revolution, concentrations of atmospheric carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) have been increasing year after year; thus, global climate change caused by greenhouse gas emissions has become a common concern in the international community (Lubbers et al., 2013; Hussain et al., 2015; Xuejun and Fusuo, 2011; Shakoor et al., 2018). CH_4 is the smallest hydrocarbon and is a potent greenhouse gas (IPCC, 2013). It is mainly produced through the decomposition of organic waste and is also one of the main causes of global warming (Zhang et al., 2015b). Although its atmospheric concentration is much lower than that of CO_2, the rate of its emission is increasing at a much higher rate. The greenhouse effect of CH_4 is 27 times larger than that of CO_2, and its current contribution to the rate of global warming is about 15% (Liu et al., 2015; Hofmann et al., 2016).

The main sources of CH_4 in the atmosphere are natural wetlands, paddy fields, termites, ruminants, fossil fuel production processes, waste disposal and shallow water lakes (Karakurt et al., 2011; Musenze et al., 2014). It is estimated that 40 to 52% of

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yearly CH_4 emissions are from soil, and farmlands are an important source of greenhouse gas emissions from soil ecosystems (Montzka et al., 2011; Linquist et al., 2012). Paddy fields are the main source of farmland soil CH_4 emissions. Globally, the total yearly amount of CH_4 emissions from paddy fields is between 35 and 150 Tg, which accounts for about 12% of global CH_4 emissions (Le Mer and Roger, 2001; Hofmann et al., 2016). Rice is one of the most important cereal crops in China, and its planting area accounts for 17.8% of the world total. CH_4 emitted from paddy fields accounts for 17.9% of the total CH_4 emissions in China, and is the second largest source after livestock farming (Yan et al., 2009; Hoben et al., 2011). Therefore, the studies on CH_4 emission characteristics are of great significance for the reduction of CH_4 emissions from paddy fields.

There are many factors affecting CH_4 emissions from farmland, which can be summarized into three categories: climatic factors such as temperature, precipitation, solar radiation intensity, and atmospheric CO_2 concentration (Walter et al., 2001); soil factors, such as soil organic matter and oxide content, texture, pH value (Wagner et al., 2005); Human factors include water management, application of organic manure and fertilizer amounts, types and dosage (Zou et al., 2005; Mohanty et al., 2017). Straw-return is one of the most important measures for improvement of farmland ecological environment. It improves soil structure, increases soil organic matter content and promotes a virtuous circle of ecosystem health (Lou et al., 2011; Nyamadzawo et al., 2015; Wang et al., 2017). In the rice paddy ecosystem, the use of straw provides a rich substrate for soil methanogenic bacteria, which significantly promotes CH_4 emissions from rice paddies (Ma et al., 2009; Qin et al., 2016; Koga and Tajima, 2011). The choice of straw type (Wassmann et al., 2000), adjustment of the application pattern (Zhu et al., 2014) and application time (Zhang et al., 2015a) maintain soil fertility while reducing CH_4 emissions to a certain extent. Bao et al. (2016) demonstrate that addition of nitrate influences transcriptional and functionally active methanogens, and can alleviate CH_4 production associated with a straw amendment in paddy soil incubations; this could occur through competition for common substrates between nitrate-utilizing organisms and methanogens (Shakoor et al., 2016). Hu et al. (2016) indicated that returning wheat straw prior to rice transplantation significantly increased seasonal CH_4 emissions during both the rice seasons and wheat seasons, compared to no straw-return. At the same time, annual CH_4 emissions were lower under ditch-buried wheat straw-return than that under wheat straw returned with rotary tillage and plowing. Zhang et al. (2015c) determined that compared with straw removal, straw-return significantly increased annual CH_4 emissions by 35.0%, annual GWP by 32.0%, and annual GHGI by 31.1%.

As a typical traditional agricultural region of China, the Chao Lake Basin has problems with excessive fertilizer application, low fertilizer utilization rates, and serious agricultural non-point source pollution. Beyond that, Chaohu’s agriculture had not implemented continuous straw-return before 2013. Therefore, there are no systematic studies on the effect of straw-return on production, oxidation, and emission of CH_4 in the subsequent rice cropping season. To define the contribution of straw-return and fertilization practices on CH_4 emissions, this experiment was designed to test plots with and without the application of wheat straw. The CH_4 emission flux was observed in situ throughout the following rice season; the CH_4 production potential and soil oxidation potential of paddy soils were studied to explore the effect of wheat straw application on CH_4 production, oxidation, and emission during the subsequent rice season.

Materials and methods

Experimental site

The field experiments were performed on the rice crop from a rice-wheat rotation system at the Chaohu Experiment Station of Anhui Agriculture University, Hefei, China between June 2013 and October 2014 (117° 40’ E, 31° 39’ N, altitude 17 m). The research site is situated in the north subtropical humid monsoon climate zone and it is within the Chao lake water regulation zone, which makes it suitable for the growth of crops (*Fig. 1*).

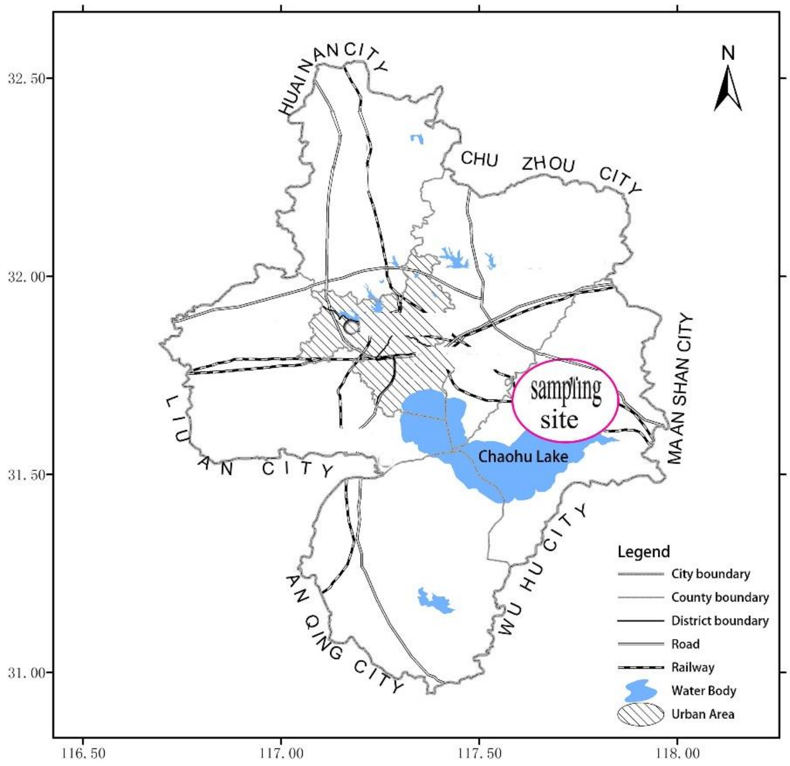

Figure 1. *Localization of the study area in Chaohu, China. (ArcGIS, ArcMap 10.2 version; http://www.esri.com/arcgis/about-arcgis; http://desktop.arcgis.com/en/arcmap/)*

The subtropical monsoon climate prevailed in the area with mean annual temperature and precipitation of 16.8 °C and 1358.3 mm, respectively. From 1986 to 2005, the mean seasonal temperature was 16.29 °C, which was similar to our findings (Shi et al., 2008). The soil type at the monitoring site is clay loamy soil with maximum water holding capacity. The specific physical and chemical properties of soil (0-10 cm) were: pH (H_2O) 6.18; Organic matter 23.64 g kg^{-1}; Total nitrogen 1.30 g kg^{-1}; clay content (particle size < 0.01 mm) 490 g kg^{-1}, respectively.

Treatment design and field management

The two phases of experiments included effects of different fertilization techniques and tillage patterns. The experiment consisted of 4 treatments, and each treatment was repeated 3 times; each experimental plot was 30 m^2, and the field was randomly sectored. Four treatments were adopted as follows:

1. Control treatment (CK): no fertilization during the rice season.
2. Traditional treatment (CT): traditional fertilization during the rice season.
3. Optimized fertilization (CO): fertilization optimized for maximum local economic output.
4. Optimized fertilization with straw-return (CO + SR): all straw produced in the wheat season is returned to the field and an additional 2 kg of the decomposing agent is applied per acre in conjunction with a 20% reduction in application of the optimized fertilizer amount.

Fertilizer application methods were application after sowing, rice base fertilizer application after seedling transplantation, and two top-dressing applications during the tillering and heading stages. The total amount of applied fertilizer and the amount of applied nitrogen for each stage are shown in *Table 1*.

Table 1. *Fertilization scheme of 2013 and 2014 rice season. (kg/hm^2)*

	Treatments	**Total amount of the fertilizer**			**Base fertilizer**			**Tillering fertilizer**	**Panicle fertilizer**	
		N	P_2O_5	K_2O	N	P_2O_5	K_2O	N	N	K_2O
Rice season	CK	0	0	0	0	0	0	0	0	0
	CT	180	67.5	67.5	67.5	67.5	67.5	67.5	45	0
	CO	225	67.5	120	90	67.5	84	90	45	36
	CO + SR	180	54	96	72	54	67.2	72	36	28.8

Management of all the fields was maintained at the same times, and in accordance with local routine management practices. During the rice growing period, irrigation was needed between 4 and 5 times. Fields were irrigated to a depth of 6-7 cm, 1 to 2 days prior to fertilizer application and at the end of the land sunning period. The amount of irrigation water was 822.7 mm. The rice crop was planted in May and harvested in early October. Rice plants were transplanted to the main field at a density of 20 hills per m^2 on May 25/26 and harvested on October 10/11 for the entire experimental period. The rice paddy field was regularly weeded to ensure the healthy growth of rice under normal conditions. Data was gathered for the individual cropping periods, which were June 13^{th,} 2013 – September 27^{th,} 2013 and June 21^{st,} 2014 – October 10th 2014, respectively (*Table 2*).

Table 2. *Fertilization scheme for the 2013 and 2014 rice seasons. (kg/hm^2)*

Management practice	**2013**	**2014**
Field management		
Transplanting	Jun 13	Jun 20
Basal fertilizer	Jun 12	Jun 21
Tilling fertilizer	Jun 28	Jul 8
Panicle initiation fertilizer	Jul 27	Aug 5
Harvest	Sept 27	Oct 10

Gas sampling and measurements

A static closed chamber was constructed with polyester material, and was used to measure the CH_4 fluxes (Chadwicka, 2014; Roche et al., 2016) and the sample box was made of 5 mm thick transparent organic glass. The sampling box was divided into upper and lower layers. In 2013, the box height was 1.2 m, and the dimensions of each layer were 0.5 m × 0.5 m × 0.6 m with an area of 0.25 m^2. In 2014, the height of the box is 1.0 m, and the dimensions of each layer were 0.5 m × 0.5 m × 0.5 m with an area of 0.25 m^2 (0.5 m × 0.5 m). A single-layer static box was used during the early growth stage, and a two-layer static box was used later when plants were higher than 0.6 m. The chamber was equipped with a thermometer on the top to measure the internal temperature. When taking samples, the chamber was placed into the base of the tank above. CH_4 gas samples were taken between 8:00 am and 11:00 am from the paddy fields, and 60 mL of the gas sample was extracted with a syringe 0, 10, 20, and 30-min interval after closing the chamber, respectively. At the same time, air temperature surface soil temperature and soil Eh at depths of 0, 5 and 10 cm were also measured. Daily precipitation and temperature were monitored with an automatic meteorological station approximately 500 m from the experimental plots.

Gas samples were collected from the first day of rice cultivation, and once every seven days thereafter. Samples were collected more frequently during the fertilization and land sunning periods. In the rice season, the conventional sampling frequency was every 3 to 5 days, but sampling was occasionally delayed in the event of precipitation. From 1 to 9 days after fertilization, data were collected every two days. In addition, sample analysis frequency increased during the land sunning period.

Data processing and statistical analysis

The samples were analyzed within 24 h. Gas chromatography (GC) was performed on a 450-GC system (Bruker Daltonics Inc., U.S.A.). CH_4 was detected by a flame ionization detector, and the emission flux was calculated using the following formula: $F = \rho \times V / A \times dc / dt \times 273 / T$, where F is the emission flux in units of $mg.m^{-2}.h^{-1}$ for CH_4, ρ is the density of CH_4 under standard conditions (0.714 $kg.m^{-3}$ for CH_4), V is the effective volume of the sampling chamber (m^3), A is the area covered by the sampling chamber (m^2), dc/dt is the change of CH_4 concentration ($\mu L.L^{-1}.h^{-1}$) in the sampling chamber per unit of time (positive value: gas emission; negative value: gas absorption), T is the temperature inside the chamber (K) and 273/T is the temperature impact factor. CH_4 emissions were calculated using a trapezoidal method according to the following formula: $Q = (F1 + F2) \times (t2\text{-}t1)/2\times24$, where Q is the total emission amount of CH_4 ($mg.m^{-2}$) and F1 and F2 are the corresponding emission fluxes at days t1 and t2. The temperature inside the chamber was measured simultaneously for the standardized correction of the volume of gas. Soil Eh was measured using a FJA-6 automated oxidation-reduction potential (ORP) analyzer (Nanjing Chuan-Di Instrument & Equipment CO., LTD. Nanjing, China).

All statistical analyses were performed using SPSS 17.0 (SPSS, Inc., USA) and EXCEL 2010 for Windows. Average fluxes and standard deviations of CH_4 were calculated based on data from triplicate plots. Differences in seasonal CH_4 emissions and grain yields between treatments were analyzed with two-way analysis of variance (ANOVA) and least significant difference (LSD) test. To determine correlation, Pearson's correlation test was applied at 5% level of significance. The map of study site was generated by using ArcGIS software (ArcMap 10.2 version; https://www.esri.com/en-us/arcgis/about-arcgis/overview). Finally, Origin 8.0 (Origin Lab Corporation, USA) was employed for figure preparation.

Results

Environmental conditions and crop productions

Chao Lake has a subtropical moist monsoon climate, and this region is dominated by both westerly circulation and subtropical circulation. Annual mean air temperatures were 15.8 °C and 16.3 °C for the 2013–2014 and 2014–2015 cropping years, respectively (*Fig. 2*). The respective annual precipitation values were 1065.6 mm and 1077.4 mm for the cropping years of 2013–2014 and 2014–2015, respectively (*Fig. 2*). The magnitudes and seasonal patterns of soil temperatures correlated with air temperatures during the two experimental years.

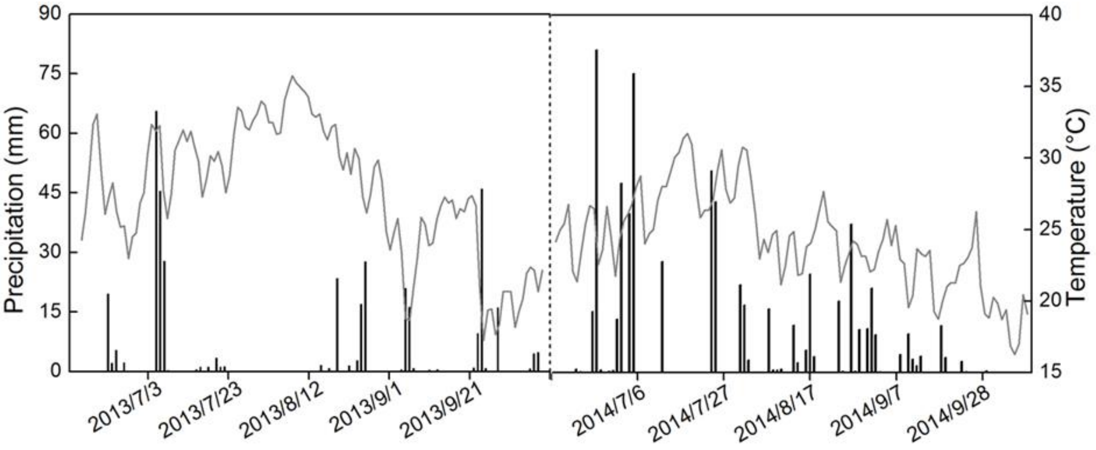

Figure 2. *Daily precipitation and mean air temperatures during the three rice growing seasons from 2013 to 2014*

CH_4 emissions during the rice-growing season

CH_4 emissions not only correlated with the influencing factors of substrate, environment, and fertilization but were also highly correlated with rice growth stage. Over the whole rice seasons from 2013 to 2014, the variable seasonal pattern of CH_4 emissions from rice paddies consisted of “three peaks”, as shown in *Figure 3*.

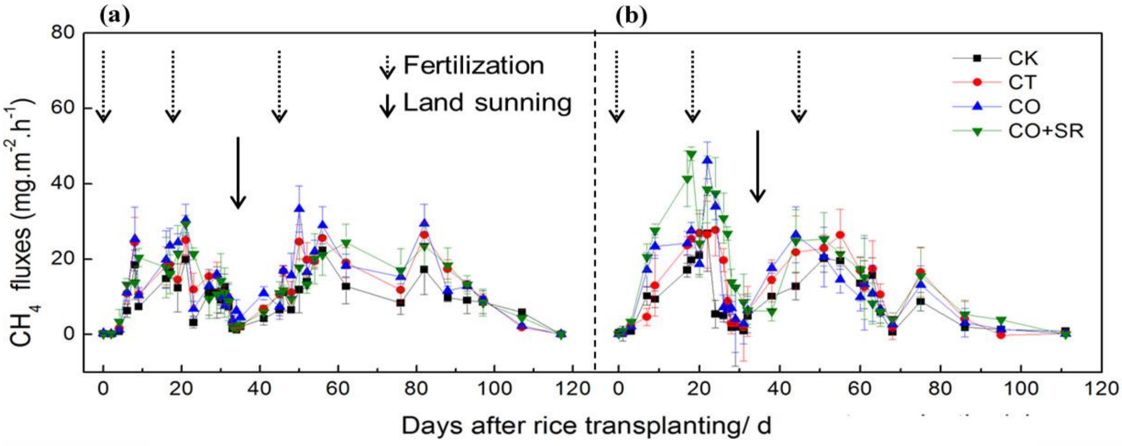

Figure 3. *Variation in methane emissions during the rice growing seasons of (a) 2013 and (b) 2014. The vertical bars represent standard errors of the means (n = 3)*

Due to the dry farming system in non-rice growing years, the soil environment had shifted to a state of oxidation; thus, CH_4 emissions did not appear during the early rice transplantation or with the latter fertilizer application. After a period of flooding, the soil state gradually shifted to a reducing environment, which directly resulted in gradual decreases in soil Eh and gradual increases in CH_4 emissions.

The first CH_4 emission peak appeared on the 22nd day after transplanting under the combined effects of fertilizer, substrate, and environmental factors. The order of CH_4 emission fluxes under each treatment were CO > CO + SR > CT > CK in 2013, and CO + SR > CO > CT > CK in 2014. During the drying of paddy fields in the sunshine (Days 23-31 in 2013 and days 27-35 in 2014), there were reductions in CH_4 production from methanogens, and the activity of methane-oxidizing bacteria was increased; thus, the CH_4 emissions from rice fields were very low during this period. After the re-flooding, the CH_4 emission flux of CK, CT, CO, and CO + SR increased slowly. With the application of panicle fertilizer on the 45th day, CH_4 emissions reached a high level and attained its second peak between days 50 and 55 both in 2013 and 2014. After 5 days of drying the paddy fields in the sunshine, the CH_4 emission flux decreased to a low level, but there were still measurable amounts of CH_4 emissions, and sixth-day observations showed a recovery of CH_4 emission fluxes. This may have been due to precipitation during the latter stages of drying. Even after the disappearance of field water, the soil retained a certain amount of moisture, and the added rainfall also contributed to the CH_4 emission flux recovery. The third CH_4 emission peak appeared during the blooming stage. The peak of CH_4 emissions was higher from CT, CO and CO + SR relative to CK, which was because the CK treatment lacked fertilizer; thus, the CT, CO, and CO + SR emission peaks were mainly caused by the input of nitrogen fertilizer.

Annual CH_4 emissions and global warming potential

The different fertilization treatments of the Chao Lake Basin experimental plots resulted in differential CH_4 emission fluxes (*Fig. 4*).

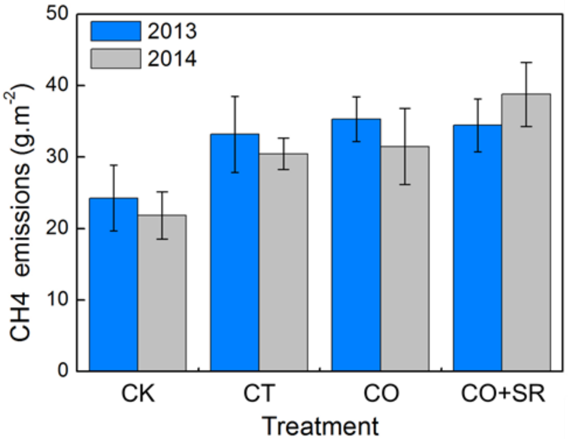

Figure 4. *CH_4 emissions under different fertilizer practices throughout the experimental period in 2013 and 2014. The vertical bars indicate standard errors of three replicates*

The total seasonal CH_4 emissions from all fertilization treatments were higher than the total CH_4 emissions from the CK treatment, indicating that application of chemical

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nitrogen fertilizer could increase CH_4 emissions from single-cropping paddy fields. Different fertilization and straw-return practices had different effects on the seasonal CH_4 emission accumulations; the seasonal CH_4 emission accumulation was calculated and significant differences were determined ($P < 0.05$) by LSD analysis of significant differences (*Table 3*). During the entire 2013 rice growth period, the total CH_4 emissions in CO treatments were the highest. The total CH_4 emitted was 35.3 ± 3.3 gm^{-2} (*Table 3*), which indicated that there was a certain relationship between CH_4 emission and the applied amounts of chemical nitrogen fertilizer. Compared with the CK treatment, CH_4 emissions from the CT, CO, and CO + SR treatments were increased by 36.6%, 45.8% and 42.0%, respectively. Although the optimized fertilization treatment CH_4 emission was 3.8% higher than that of the Optimized fertilization plus straw-return treatment, the difference in CH_4 emissions was not significant. During the entire 2014 rice growing period, the total CH_4 emitted from the CO + SR treatment was the highest and amounted to 38.8 ± 4.5 g m^{-2} (*Table 3*).

Table 3. *Average annual yields and emissions of CH_4 expressed as CO_2-equivalents for the rice growing seasons in 2013 and 2014*

Year	Treatment	CH_4 emission ($g \cdot m^{-2}$)	Rice yield ($kg \cdot hm^{-2}$)	Total E-CO_2 ($kgCO_2 \cdot hm^{-2}$)
2013	CK	24.3 ± 4.6a	5888.9 ± 146.9b	6063.6
	CT	33.2 ± 5.4a	7250.0 ± 127.3ab	8292.3
	CO	35.3 ± 3.3b	7611.1 ± 111.1a	8826.7
	CO + SR	34.5 ± 3.7b	7333.3 ± 364.3b	8613.7
2014	CK	21.8 ± 3.4c	5966.7 ± 135.6c	5456.6
	CT	30.4 ± 2.2b	8376.7 ± 189.6b	7612.5
	CO	31.5 ± 5.3a	8670.3 ± 111.1a	7870.5
	CO + SR	38.8 ± 4.5a	8273.4 ± 140.1a	9690.3

Data in the first two columns are expressed as Means ± SD; different letters within the same column denote significant differences in variable means among treatments over the 2013-2014 seasons based on the LSD multiple-range test ($P < 0.05$).

Compared with the CK treatment, the CT, CO, CO + SR treatments had increases in CH_4 emissions of 42.0%, 48.5%, and 80.1%, respectively. The maximum CH_4 fluxes were different between the two the rice growing season. The CH_4 emissions in 2014 from CK, CT and CO treatments showed a similar pattern as those of 2013, but were all lower than those in 2013; this was probably due to the inter-annual climate, rice growth conditions and other variability's, which led to differences in CH_4emission flux. There was a significant increase inCH_4 emission in the treatment of straw returning to the field in 2013 and 2014. The cumulative 2013 CH_4 emitted from straw-return was 1.42 times that of CK treatment, which increased to 1.80 in 2014; this indicated that straw-return increased the CH_4 emissions in Chao Lake rice paddies.

Effects of soil Eh during the rice-growing season on CH_4 emissions

Since methanogenesis requires substrates and extreme reducing conditions, sufficient methane-producing substrates and a suitable methanogenic growth environment are prerequisites for CH_4 production. During the early stage of rice growth, soil Eh was still high (*Fig. 5*), which hindered CH_4 production in the soil, resulting in almost no CH_4 production after the first two treatments (*Fig. 3*). During the first four days after transplanting, CH_4 fluxes remained at 0 mg m^{-2} h^{-1} and began to increase rapidly for 8 days starting on day 6 to reach the first peak. These changes correlated well with increases in soil Eh. In the middle stage of rice growth, the soil Eh had decreased levels suitable for methanogenesis, and there was un-degraded straw remaining from the straw-return CO + SR treatment; this provided a rich methanogenic substrate and appropriate environment for CH_4 production, thereby promoting the production of CH_4 in paddy fields.

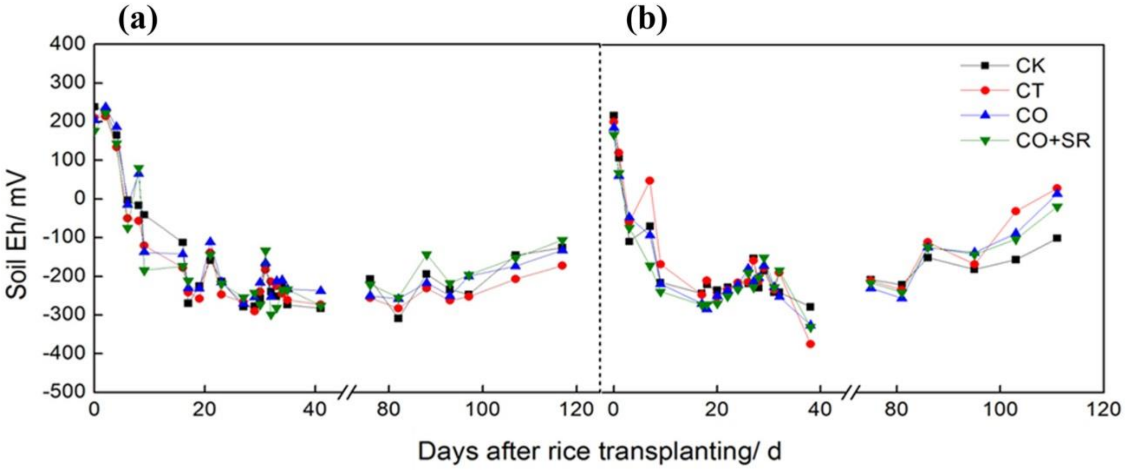

Figure 5. *Seasonal dynamic of soil Eh during the rice growing seasons of (a) 2013 and (b) 2014*

During the first four days after transplanting, CH_4 fluxes remained at 0 mg m^{-2} h^{-1} and began to increase rapidly for 8 days starting on day 6 to reach the first peak. These changes correlated well with increases in soil Eh. In the middle stage of rice growth, the soil Eh had decreased levels suitable for methanogenesis, and there was un-degraded straw remaining from the straw-return CO + SR treatment; this provided a rich methanogenic substrate and appropriate environment for CH_4 production, thereby promoting the production of CH_4 in paddy fields. The CH_4 production potential of the two treatments increased markedly from 35 to 66 days after transplanting. During the later stage of rice growth, soil Eh was not the limiting factor for CH_4 production. With the consumption of straw, CH_4 production potential gradually decreased and eventually stabilized. The straw mulch did not decompose because of the absence of straw organic matter supplement until the soil Eh decreased to levels suited for CH_4 production. The correlation between CH_4 emission flux and soil Eh is shown in *Table 4*.

Table 4. *Correlations of the variations in CH_4 emission flux with soil Eh*

Treatment	CK	CT	CO	CO + SR
E_h	-0.536	-0.652*	-0.571*	-0.736**

*At the $P > 0.05$ level significantly correlated (bilateral). **At the 0.01 level significantly correlated (bilateral)

Effects of temperature on CH_4 emissions during the rice-growing season

Temperature affects both CH_4 production and emission, and the CH_4 emission flux from paddy field closely correlates with air and soil temperature. Correlation analysis was performed for CH_4 emission flux and surface temperatures at depths of 5 and 10 cm during the sampling period, and the correlation coefficients are shown in *Table 5*.

Table 5. *The correlation of the variations of the CH_4 emission flux with soil temperature*

Treatment	Soil surface temperature	5 cm depth temperature	10 cm depth temperature
CK	0.074	0.320*	0.169
CT	0.128	0.308*	0.116
CO	0.290*	0.335**	0.135
CO + SR	0.270*	0.312*	0.196

*Significantly correlated at the P > 0.05 level (bilateral); **significantly correlated at the P > 0.01 level (bilateral)

CH_4 emission fluxes were significantly correlated with soil temperatures at 5 cm depth ($P < 0.05$) for all treatments and with surface temperatures (0 cm depth) for the CO and CO + SR treatments; the land surface temperatures failed to correlate with CH_4 emission fluxes for CK and CT, whereas temperatures at a depth of 10 cm failed to correlate with CH_4 emission fluxes for any treatments. A large number of studies have shown that CH_4 emissions from rice fields are affected by many factors, such as soil properties, rice growth, tillage measures and climatic factors. Under the conditions of continuous flooding and with an adequate supply of organic matter, soil Eh was non-limiting and changes of CH_4 emission fluxes were mainly affected by the factors such as soil temperature. Compared with the surface temperatures and soil temperatures at a depth of 10 cm, CH_4 fluxes were more closely related to soil temperature at 5 cm depth; this may be because the soil CH_4 activity mainly occurred in 0 ~ 5 cm soil layer and decreased significantly thereafter. In addition, straw mulching had a significant effect on the surface temperature, which could increase the soil temperature at 5 cm to a certain extent, thus promoting the activity of methane-producing bacteria.

Relationship between CH_4 flux and soil temperature at 5 cm depth

The soil temperature at 5 cm depth was the main environmental factor affecting the CH_4 flux in paddy fields, and the regression equation was obtained by regression analysis between the CH_4 flux and the corresponding of 5 cm soil temperature values: CK: $Y = -17.30573 + 0.84321X$ ($R^2 = 0.14732$, $P < 0.05$); CT: $Y = -24.27339 + 1.19103X$ ($R^2 = 0.16676$, $P < 0.01$); CO: $Y = -28.48551 + 1.35211X$ ($R^2 = 0.17485$, $P < 0.05$); CO + SR: $Y = -24.55386 + 1.17238X$ ($R^2 = 0.14732$, $P < 0.05$), where x is the temperature and y is the CH_4 emission flux (*Fig. 6*). According to the equation, CH_4 emission fluxes were positively correlated with temperature, and the methanogenic activity of the 0~5 cm soil layer was higher than that of deeper soils.

Discussion

Numerous latest studies have shown that application of nitrogen fertilizer positively correlates with CH_4 emissions from agricultural soils (Parkin and Hatfield, 2013;

Halvorson et al., 2014). CH_4 emission fluxes from rice cropping fields ranged between 35.3 and 38.8 g m^{-2} over the experimental period, which agreed with results from previous studies (25.9 gm^{-2} to 41.9 gm^{-2}) conducted in different regions (Ku et al., 2017; Aulakh et al., 2001; Hu et al., 2016). In a terrestrial environment, there are numerous factors affecting CH_4 emissions such as denitrification, nitrification, chemodenitrification, heterotrophic nitrification, codenitrification and oxidation of ammonia; these processes are directly affected by the application of nitrogen fertilizer in the soil (Shurpali et al., 2016; Zhang et al., 2016; Luo et al., 2016; Shakoor et al., 2016). The results of this study also support this conclusion. In the same way, we analyzed the effects of nitrogen application on CH_4 emissions and emission peaks during the rice cropping seasons. In this study, the CK control treatment did not use nitrogen fertilizer application, and both its seasonal and annual CH_4 accumulated emissions were significantly lower than the other fertilization treatments. Seasonal CH_4 emissions fluxes observed by Ku et al. (2017) averaged a very low 15 gm^{-2} with nitrogen fertilizer applied at 150 kg ha^{-1}. Similarly, with reduced nitrogen fertilizer application to different agricultural fields (Molodovskaya et al., 2011; Ussiri et al., 2009; Deng et al., 2015), lower CH_4 emissions fluxes were reported. This study found that increasing the application of nitrogen fertilizer could promote CH_4 emission from soil into the atmospheric environment. This study confirmed that the straw returning significantly affected the CH_4 emission in the soil, whereas reduced application of nitrogen fertilizer can decrease CH_4 emissions. In 2014 rice growing period, the total CH_4 emitted from the CO + SR treatment was the highest among all the treatments and amounted to 38.8 gm^{-2} (*Table 3*). Similarly, previous studies also reported that the application of straw retuning can significantly increase CH_4 emissions (Wang et al., 2017, 2016, 2015).

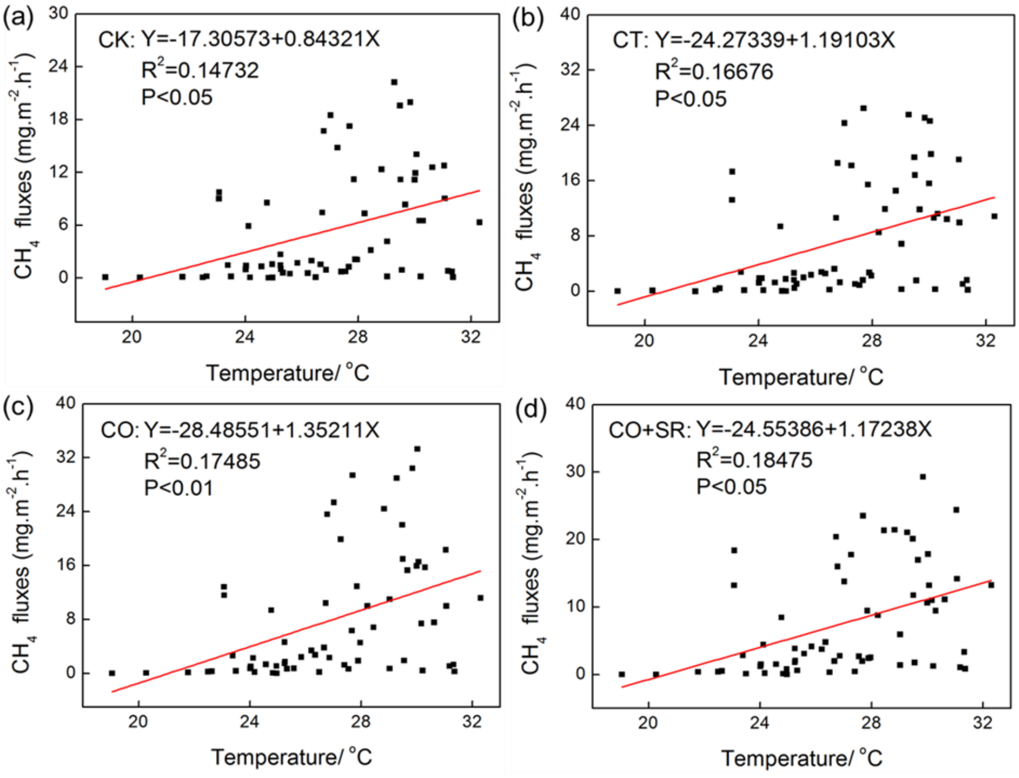

Figure 6. *Relationship between methane fluxes and soil temperatures of the 5 cm soil layer, (a) CK, (b) CT, (c) CO, (d) CO + SR*

Generally, CH_4 is emitted during soil denitrification and nitrification processes (Shakoor et al., 2016; Smith et al., 1997), which are highly related to soil temperature (Smith et al., 1997; Sun et al., 2016; Aulakh et al., 2001); thus, soil temperature can greatly influence CH_4 emissions. Increased emissions of CH_4 as soil temperature increased from 25 °C to 30 °C showed that production of CH_4 was sensitive to soil temperature (Liu et al., 2017). In this study, the average soil temperature was 15.6 °C with a range of -3.1 °C to 34.5 °C. Maximum CH_4 emissions were observed at 27.5 °C, which was similar to results from recent studies (Maljanen et al., 2017; Liu et al., 2017). Gaihre et al. (2013) also studied that higher temperature could responsible for elevated of CH_4 emission, which was further increased by the application of straw returning.

Soil Eh is a key factor of CH_4 emission from paddy fields and it does not only affect methanogens activities but also involved in gas transformation through the aerenchyma (Le Mer and Roger, 2001). At a lower value of soil Eh, methanogens activities and formation of aerenchyma were increased (Kludze et al., 1993). Significant emissions of CH_4 occur once soil Eh is < -100 mV (Hou et al., 2016; Gaihre et al., 2013). So, the emission of CH_4 is negatively correlated to soil Eh. In our findings, the value of soil Eh was very high during the early stage of rice growth (*Fig. 5*), which delayed CH_4 production and emissions, which was similar to previous recent studies (Charles et al., 2017; Xu et al., 2017; Gaihre et al., 2013; Wang et al., 2017).

Conclusions

The experimental results of two - year farmland in Chao lake single-cropping rice fields showed that: CH_4 emission pattern from rice field consisted of “three peaks”, which appeared before land sunning and after the re-flooding period; the seasonal variation pattern was consistent. CH_4 seasonal emissions were lowest under the CK treatment for both years and were highest under the CO treatment during 2013 and the CO + SR treatment in 2014. The CO + SR treatment may have produced less CH_4 in 2013 due to a large number of stalks initially returned to the field, resulting in an excessive C/N ratio which inhibited soil respiration and affected microbial activities. A year later, the physical and chemical properties of the land had reached a balance, which could better reflect the effect of straw return on CH_4 emissions. The CO + SR treatment increased the total CH_4 emissions by 42.0% and 80.1% in 2013 and 2014, respectively. There was a significant correlation between CH_4 emissions from CO + SR treatment and soil temperatures at depths of 0 and 5 cm, but not at 10 cm. In addition, there was a significant negative correlation between CH_4 emissions and soil E_h. We estimated total CH_4 emissions from the rice growing periods of the two years, and emissions of the different treatments were CO + SR > CO > CT > CK. Straw return to soil can improve soil fertility, increase rice yield, but also increases the CH_4 emissions from rice fields. Therefore, it is necessary to consider the problem of straw returning from no-tillage paddy fields to effectively realize the carbon sequestration potential and reduce the emission of greenhouse gases.

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