Nutr Cycl Agroecosyst (2017) 107:369–380
DOI 10.1007/s10705-017-9838-x

ORIGINAL ARTICLE

Land use legacies and nitrogen fertilization affect methane emissions in the early years of rice field development

Rui Shao · Ming Xu · Renqiang Li · Xiaoqin Dai · Lixiang Liu · Ye Yuan · Huimin Wang · Fengting Yang

Received: 27 June 2016 / Accepted: 7 March 2017 / Published online: 11 March 2017

Abstract Methane (CH_4) emissions are critical to greenhouse gas (GHG) management in agriculture, especially in areas growing rice (*Oryza sativa*). However, studies on CH_4 emissions and the nitrogen (N) fertilization effect in new rice fields in subtropical regions are still scarce. In this study, we designed a split-plot field experiment in Jiangxi Province, southern China, to examine whether land-use legacies and N fertilization would influence CH_4 emissions. Using static chambers and gas chromatography, we measured CH_4 fluxes in a newly developed rice paddy and a 10-year-old rice paddy. We also measured climatic factors and soil chemical and physical properties to match the flux measurements. The results showed that annual CH_4 emissions in the new rice plots were significantly lower than in the old rice plots regardless of N fertilization. Annual CH_4 emissions increased with the land-use years of rice paddies, following the order of 1 year < 2 years < 3 years < 10 years. N fertilization significantly decreased CH_4 emissions by 36.9% in the first year after the new rice plots were developed, whereas it had no significant effects on

---

R. Shao · M. Xu (✉) · R. Li · X. Dai ·
L. Liu · Y. Yuan · H. Wang · F. Yang
Key Laboratory of Ecosystem Network Observation and Modeling, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, 11A Datun Road, Beijing 100101, China
e-mail: mingxu@igsnrr.ac.cn

R. Shao
e-mail: ruishao1010@gmail.com

R. Li
e-mail: renqiangli@igsnrr.ac.cn

X. Dai
e-mail: daixq@igsnrr.ac.cn

L. Liu
e-mail: liulixiang1982@126.com

Y. Yuan
e-mail: 276292864@qq.com

H. Wang
e-mail: wanghm@igsnrr.ac.cn

F. Yang
e-mail: yangft@igsnrr.ac.cn

R. Shao · M. Xu
University of Chinese Academy of Sciences,
Beijing 100049, China

M. Xu
Department of Ecology, Evolution and Natural Resources,
Center for Remote Sensing and Spatial Analysis, Rutgers,
The State University of New Jersey, New Brunswick,
NJ 08901, USA

Y. Yuan
College of Life Sciences, Anhui Normal University,
Wuhu 241000, China

CH_4 emissions in the old rice plots or the new rice plots in the second and third years. The results suggest that land-use legacies have significant effects on CH_4 emissions and may influence the N fertilization effect on CH_4 emissions in rice fields in subtropical regions. The findings suggest that land-use legacies should be considered in managing and estimating GHG emissions in rice-growing regions.

Keywords Land use change · Land-use years · CH_4 · Subtropical region · China

Introduction

Atmospheric CH_4 is recognized as one of the most important greenhouse gases (GHG) due to its high global warming potential (GWP) (34 times greater than CO_2 at the per molecule level in a 100-year time horizon) (Myhre et al. 2013). CH_4 contributes to approximately 20% of the total global warming forcing (Schulze et al. 2009), and it is projected that CH_4 emissions may increase by as much as 60% by 2030 (Smith et al. 2007). Paddy rice cultivation is a major source of global CH_4 emissions, contributing to approximately 11% of the total global CH_4 emissions (Smith et al. 2014). To meet food demands for the world's growing population, rice production has to increase by 60% in the next few decades (Cassman et al. 1998). As the largest rice producer in the world, China produces approximately 36% of the global rice with 23% of the world's total rice planting area (Dong et al. 2011).

Double rice cropping systems are widely adopted in southern China, representing the dominant crop system in the region (Yang et al. 2010). In the past several decades, new rice fields have been widely developed in southern China due to rapidly increasing rice demand from population growth. Land-use legacies arising from land-use change could impact ecosystem carbon and nitrogen cycles in various ways, such as changing soil nutrient pools (McGrath et al. 2001; Zhang and He 2004), modifying soil microbial communities (Chu et al. 2009) and altering GHG emissions (Eusufzai et al. 2010; Nishimura et al. 2011). Several studies showed that CH_4 emissions were much lower in new rice fields recently converted from uplands than old rice fields (Eusufzai et al. 2010; Nishimura et al. 2011). Multi-year studies on CH_4 emissions in new rice fields are necessary to investigate the lagged effects of land-use change (Hatala et al. 2012) and are important for improving GHG inventories. However, long-term studies on CH_4 emissions in new rice fields converted from upland cropping systems are still scarce, especially in subtropical regions.

CH_4 emissions in new rice fields can also be affected by many other factors, such as water and fertilization management practices. As an effective way to increase rice production, intensive management practices with high nitrogen inputs through fertilization have been adopted widely in south China. Nitrogen fertilization generally leads to greater N_2O emissions (Zou et al. 2005; Shang et al. 2011). However, N fertilization can also reduce CH_4 emissions (Banger et al. 2012; Linquist et al. 2012) and thus decrease the overall GHG of CH_4 and N_2O expressed as carbon dioxide equivalents (Dong et al. 2011). However, there are no consistent conclusions on the net effects of N fertilization on CH_4 emissions (Dong et al. 2011; Shang et al. 2011; Banger et al. 2012). Some studies showed that N fertilization could increase CH_4 emissions in rice paddies (e.g., Schimel 2000; Yang et al. 2010), while others have reported a significant decrease (approximately 30–50%) (Dong et al. 2011; Xie et al. 2010) or no effects (Lindau et al. 1991; Wang et al. 2012; Pittelkow et al. 2014).

Given the importance and complexity of the N fertilization effect (NFE) on CH_4 emissions in rice paddies, numerous studies have been conducted (e.g., Bodelier et al. 2000a; Bodelier and Laanbroek 2004; Banger et al. 2012; Pittelkow et al. 2014). However, most studies have focused on N fertilization per se, such as N form and application rates (e.g., Bodelier et al. 2000b; Cai and Mosier 2000; Dong et al. 2011). Few have examined how site-specific environmental factors affect the NFE on CH_4 emissions, except for Banger et al. (2012), who reported that the NFE on CH_4 emissions was closely related to water management activities in rice paddies. Perring et al. (2016) suggested that land-use legacies could alter system dynamics by modulating contemporary environmental changes in biogeochemical processes, such as the effects of N addition on the carbon cycle. Therefore, the objectives of the current study were to examine if

(1) land use legacies affect CH_4 emissions, if (2) N fertilization affects CH_4 emissions, and if (3) land use legacies affect the NFE in rice fields in subtropical China.

Methods

Site description

The experiment began in the late rice season in 2012 at the Qianyanzhou Ecological Research Station (26°44′46″N, 115°04′05″E), which is located in Jiangxi Province in south China. The region experiences a monsoon climate, with a dry season typically extending from July to September and a rainy season from April to June. According to meteorological data, the mean annual temperature is 18.0 °C, and the mean annual precipitation is approximately 1500 mm. The double rice cropping system is the main cropping system in this region. The soil is a typical red soil and is classified as Ultisols in USDA soil taxonomy. The parent material of the soil consists of red sandstone and sandy conglomerate. The topsoil consists of 17% clay (<0.002 mm), 25% silt (0.002–0.02 mm) and 56% sand (0.02–2 mm). The soil pH was 4.97, and the bulk density was 1.29 g/cm^3.

Experimental design

We designed an experiment with four treatments in a split-block design with four replicates: old rice paddy with fertilization (OF), old rice paddy without fertilization (OUF), new rice paddy with fertilization (NF), and new rice paddy without fertilization (NUF). The old rice paddy was less than 100 m away from the new rice paddy. The old rice paddy had been cultivated continuously for 10 years before the experiment started. The new rice paddy was converted from a 5-year plantation of oil tea camellia (*Camellia oleifera*). The rice paddies had been cultivated following a conventional double-rice cropping system with an early rice season from April to July, a late rice season from July to November, and a fallow season normally from November to the following April. Both the late and early rice were transplanted at a density of 235,000 hills/ha with a spacing of 25 cm by 17 cm. Each plot had an area of 10 m × 10 m. The fertilization rate was 180 kg N/ha in each rice growing season, which is a typical rate for the study area. As a common practice for double rice systems, fertilizers were applied twice during each growing season. Compound fertilizers ($N:P_2O_4:K_2O = 15\%:15\%:15\%$) were applied first as a basal fertilizer at a rate of 108 kg N/ha at the time of rice transplanting, which was May for the early rice season and July for the late rice season. In the early-tillering stage of rice growth, we applied urea at a rate of 72 kg N/ha to the field. All plots were under typical water management, with an intermittent irrigation regime (flooding-drainage-moist during the tillering, panicle and ripening stages, respectively).

CH_4 flux measurements

Static chambers were used to collect air samples, which were then analyzed with a gas chromatograph (GC system, 7890A, Agilent Technologies, Santa Clara, CA, USA). The chamber system is composed of two parts: a movable cylindrical steel chamber body and a fixed steel base with a diameter of 49 cm, covering an area of 0.188 m^2. The base was inserted into the soil to a depth of 15 cm for the entire rice growing season. The chamber body has a diameter of 49 cm and a height of 69 cm. At the beginning of each round of measurements, we positioned the chamber body on top of the base and used a silicon tube to seal the joint to form a closed system. Two mixing fans were installed inside each chamber for mixing the air in the chamber space. The fans were powered by batteries and positioned at the top of the chamber to minimize disturbing the boundary layer. We turned on the fans before deploying the chamber body to avoid perturbation to the chamber pressure when turning on the fans. We also installed a pressure balance tube at the top of the chamber. Five rice plants were included in each base frame. Gas sampling was usually carried out twice per week. After fertilizer was applied, we collected gas samples every day for approximately a week.

We calculated CH_4 fluxes by measuring CH_4 concentration increases inside each chamber when it was closed. Gas samples from the closed chamber were collected with a custom-built manifold, which consisted of a 12 mL Exetainer vial (Labco Limited, Lampeter, Ceredigion, UK), an air pump (KNF Neuberger, Inc., Trenton, NJ, USA), a pressure sensor and a solenoid valve. The pump stopped when the pressure inside the vial reached 3 bars. The vials were evacuated with a vacuum pump prior to the field measurements. Five gas samples were collected in each chamber at 10 min intervals. The first sample was collected 30 s after the chamber was closed. The measurements for each chamber were completed in approximately 40 min. The gas samples were then transported to the laboratory and analyzed using a gas chromatograph (GC System, 7890A, Agilent Technologies) equipped with an electron capture detector (ECD) and a flame ionization detector (FID). CH_4 fluxes were determined by fitting a linear regression model where CH_4 concentration was the dependent variable and sampling time was the independent variable. The following equation was used to calculate CH_4 fluxes:

$$F = \rho \cdot V/A \cdot P/P_0 \cdot T_0/(T + 273) \cdot dC_t/dt \qquad (1)$$

where F is the CH_4 flux rate (mg C/m/h), ρ is the air density (1.29 kg/m^3), V is the chamber volume (m^3), A is the base area (m^2), P is the site air pressure, P_0 is standard air pressure at sea level (101.3 kpa), T is air temperature in the chamber (°C), T_0 is a reference air temperature of 273 K, and dC_t/dt is growth rate of the CH_4 concentration in the chamber (mg C/h).

The measurement of CH_4 fluxes in the new rice paddy continued for 3 years, while that in the old rice paddy was carried out only in the first year of the 3-year study. Annual cumulative emissions of CH_4 for all treatments were directly computed from the measured fluxes by linear interpolation for the days without measurements. We quantified the NFE on CH_4 fluxes using the difference (ΔF) of CH_4 fluxes between the plots with and without fertilization. A negative NFE meant that N fertilization reduced CH_4 emissions, while a positive NFE meant that N fertilization stimulated CH_4 emissions. N fertilization-induced CH_4 emissions were defined as CH_4 emissions per kg N fertilizer and were calculated by dividing the difference of CH_4 emissions between the plots with and without fertilization by the amount of N fertilizer (Bouwman 1996; Banger et al. 2012).

Measurements of climatic factors

Air temperature inside the chamber headspace was measured with a thermocouple simultaneously with gas sampling. Soil temperature and soil moisture at a 5 cm depth were automatically recorded by an automatic data-logging system via wireless technology. Water depth in the rice fields was monitored using a steel ruler when the fields were flooded. Air temperature and precipitation were collected from an on-site automatic meteorological station adjacent to the experimental plots.

Plant biomass

Aboveground rice biomass was determined just before harvesting. The rice plants were sampled at four random subplots at an area of 1 m x 1 m in each plot. The rice plants were then separated into grain and straw, which were oven dried at 65 °C for 48 h for dry matter weight determination. Rice roots were sampled at three 1 m × 1 m subplots immediately after harvesting in each plot. The roots were washed thoroughly, oven dried and weighed.

Soil sampling and laboratory analysis

Soil samples in each plot were collected every 10–15 days during the study period to analyze the concentrations of soil nitrate (NO_3^-–N) and soil ammonia (NH_4^+–N) from both treatments. In each plot, soils were collected using a 5-cm diameter auger in the plough layer (0–10 cm) at five randomly selected locations and then mixed as one sample. All fresh soil samples were sieved through a 2.0 mm sieve and stored in a refrigerator for nutrient analysis.

Soil pH was measured with a glass electrode in a 1:2.5 soil:water solution at the end of each rice growing season. Soil NO_3^-–N and NH_4^+–N were extracted with 1 M KCl, and the filtrate was analyzed by an AA3 HR AutoAnalyzer (SEAL Analytical GmbH, Norderstedt, Germany). Soil samples for total N and soil organic C were collected at the end of each season and air-dried. Soil total N and total C contents were analyzed by dry combustion using vario MACRO cube (Elementar Analysensaysteme GmbH, Langenselbold, Germany), while soil inorganic C was analyzed using a 08.53 calcimeter (Eijkelkamp Soil & Water, Giesbeek, The Netherlands).

Statistical analysis

All data were tested for normal distribution before statistical analysis, and for data that not normally distributed, a natural logarithm transformation with certain constants were adopted. Duncan's multiple range test was used to analyze the differences of soil temperature, moisture, soil pH, soil organic carbon content and rice biomass among the treatments in different studying years. Two-way ANOVA was used to determine the main and interactive effects of land-use legacies and N fertilization on CH_4 fluxes. One-way ANOVA with Duncan's multiple range test was used to examine the differences of the NFE on CH_4 fluxes and annual CH_4 emissions among the rice plots under different land-use years. A paired-sample *T* test was used to analyze the effects of N fertilization on annual mean fluxes and annual cumulative emissions of CH_4 in rice plots under different land-use years. All statistical analyses were conducted with IBM SPSS 22.0 (IBM, New York, USA).

Results

Environmental factors

The seasonal patterns and variations of soil temperature, precipitation and soil moisture in the rice plots are shown in Fig. 1. There were no significant differences of soil temperature and soil moisture among the different treatments during the study. Annual precipitation did not vary much among the years, with values of 1639, 1587 and 1510 mm in the three years, respectively. The soil organic carbon (SOC) content in the new rice fields was significantly lower than that in the old rice fields (Table 1). There was no significant difference of SOC in the new rice paddy among the different study years (Table 1). The soil pH in the new rice paddy increased from 4.93 to 5.11 in the NF plots and from 4.85 to 5.09 in the NUF plots during the three-year study, which was markedly lower than that in OF and OUF (Table 1).

Seasonal dynamics of CH_4 fluxes

CH_4 fluxes fluctuated considerably in all treatments during the study period (Fig. 2) with similar seasonal patterns. During the rice growing seasons, CH_4 fluxes gradually increased until seasonal peak fluxes were reached at approximately 10–25 days after rice transplanting when the plots were waterlogged. After midseason drainage, CH_4 fluxes dropped sharply in all plots. Thereafter, CH_4 fluxes remained at a low level due to the alternating dry and wet cycles until rice harvest. During the fallow seasons, CH_4 fluxes were negligible, fluctuating around zero.

The effects of land-use legacies on CH_4 emissions

Land-use legacies significantly affected CH_4 fluxes (Table 2). CH_4 fluxes from the old rice plots were significantly higher than that from the new rice plots. CH_4 fluxes increased with land-use years irrespective of N fertilization, following the order of 1-year < 2-year < 3-year < 10-year (Table 3). As a result, the cumulative emissions of CH_4 were significantly lower in the new rice paddy than the old rice paddy, regardless of N fertilization and land-use years in the new rice plots (Fig. 3). Annual CH_4 emissions also increased with the land-use years of the rice plots (Fig. 3). Annual CH_4 emissions increased by 40.7 and 49.5%, respectively, in the second and third year of cultivation of the NF plots compared to the first year. In parallel, the NUF plots observed an increase of 0.8 and 39.8%, respectively, in the second and third year compared to the first year after the new rice fields were developed.

The effects of N fertilization on CH_4 emissions

N fertilization had no significant effect on CH_4 fluxes during the study period (Table 2). However, the NFE on CH_4 fluxes varied significantly with the land-use years of the rice plots (Table 3). In the first year, N fertilization significantly decreased CH_4 fluxes from the new rice plots, whereas N fertilization had no significant effects on CH_4 fluxes from the 10-year-old rice plots (Table 3). N fertilization had no significant effects on CH_4 fluxes during the second and third year of cultivation of the new rice plots (Table 3). N fertilization significantly reduced CH_4 emissions by 36.9% from the new rice plots during the first year of cultivation, whereas it had little effect on the annual CH_4 emissions from the 10-year-old rice paddy (Fig. 3). N fertilization-induced CH_4 emissions increased with the land-use years of rice plots, following the order of 1-year < 2-year < 3-year < 10-year with values of −0.21, −0.07, −0.05 and 0.13 kg CH_4/ha/kg N, respectively.

Discussion

Land-use legacies affect CH_4 emissions

The seasonal pattern of CH_4 fluxes in the present study was in line with many previous studies (Sass et al. 1992; Yagi et al. 1996; Jia et al. 2001; Xu and Inubushi

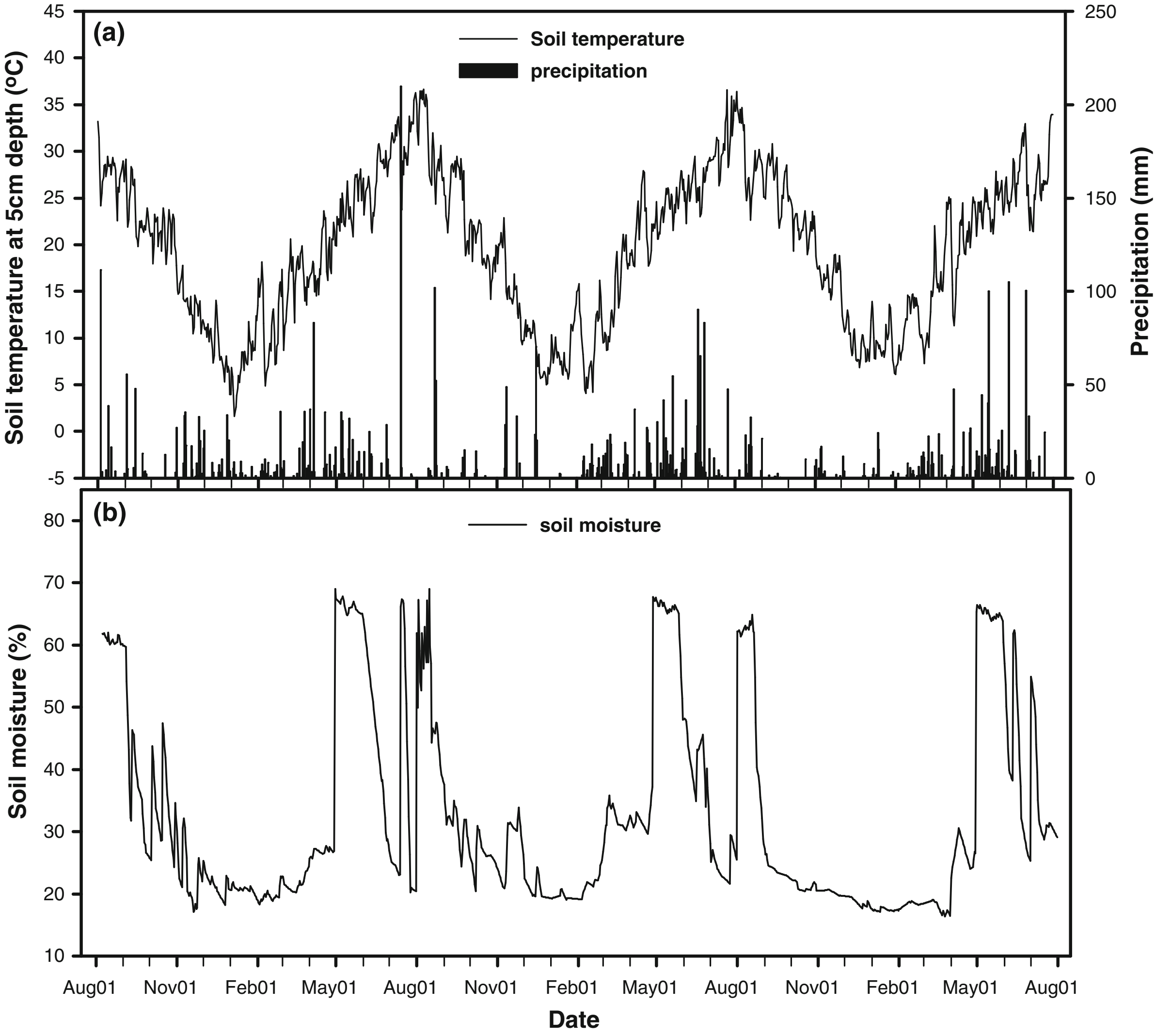

Fig. 1 Daily mean soil temperature at a 5 cm depth, daily precipitation (*upper panel*), and seasonal dynamics of soil moisture (*bottom panel*) during the study period

2004). The results showed that the seasonal pattern of CH_4 fluxes was mainly determined by water regime and rice growing stages during the rice-growing season, irrespective of the former land use history and nitrogen fertilization. Annual CH_4 emissions from the old rice plots in this study were within the range of previous studies (Feng et al. 2013). However, annual CH_4 emissions from the new rice plots were much lower than the old rice plots at the site and those under similar climatic conditions and management practices in southern China (e.g., Tang et al. 2014).

Lower CH_4 emissions from the new rice fields compared to the old rice fields were also supported by the literature. For example, CH_4 emissions from a rice paddy recently converted from upland soybean were significantly lower than that from a long-term rice paddy (19-year-old) (Eusufzai et al. 2010), and the introduction of upland crops into the double rice cultivation significantly decreased CH_4 emissions in subsequent paddy rice cultivation (Nishimura et al. 2011; Weller et al. 2016). According to previous studies, lower CH_4 emissions in the new rice fields converted from upland soils can be ascribed to land-use legacy effects, which included higher soil redox potential (Eh), lower soil organic matter content (Eusufzai et al. 2010; Nishimura et al. 2011), and the

Table 1 Climatic factors, soil properties and rice biomass (mean ± SE) under various treatments during the study period

Study Year	Land-use years	Treatments	Soil temperature[a] (°C)	Soil moisture[a] (%)	Soil pH	SOC (mg/kg)	Rice biomass (t dry matter/year)
2012–2013	10	OF	19.77 ± 0.12	34.52 ± 0.22	5.21 ± 0.03b	9.23 ± 0.31a	20.45 ± 1.09a
		OUF	19.83 ± 0.16	35.60 ± 0.17	5.33 ± 0.03a	9.42 ± 0.39a	19.06 ± 1.67a
2012–2013	1	NF	19.68 ± 0.19	34.26 ± 0.15	4.93 ± 0.01e	5.97 ± 0.01b	22.61 ± 2.29a
		NUF	19.81 ± 0.15	33.98 ± 0.21	4.85 ± 0.02e	6.02 ± 0.02b	13.35 ± 0.97b
2013–2014	2	NF	19.93 ± 0.14	33.26 ± 0.34	5.01 ± 0.02d	5.99 ± 0.02b	20.53 ± 1.38a
		NUF	19.97 ± 0.22	33.07 ± 0.46	4.98 ± 0.03d	6.03 ± 0.03b	10.97 ± 1.01b
2014–2015	3	NF	19.92 ± 0.11	34.01 ± 0.10	5.11 ± 0.03c	6.42 ± 0.03b	21.97 ± 2.08a
		NUF	19.97 ± 0.20	33.87 ± 0.16	5.09 ± 0.03c	6.01 ± 0.03b	12.21 ± 1.28b

[a] No significant differences among the treatments

Different letters in each column indicate significant differences at the $p < 0.05$ level

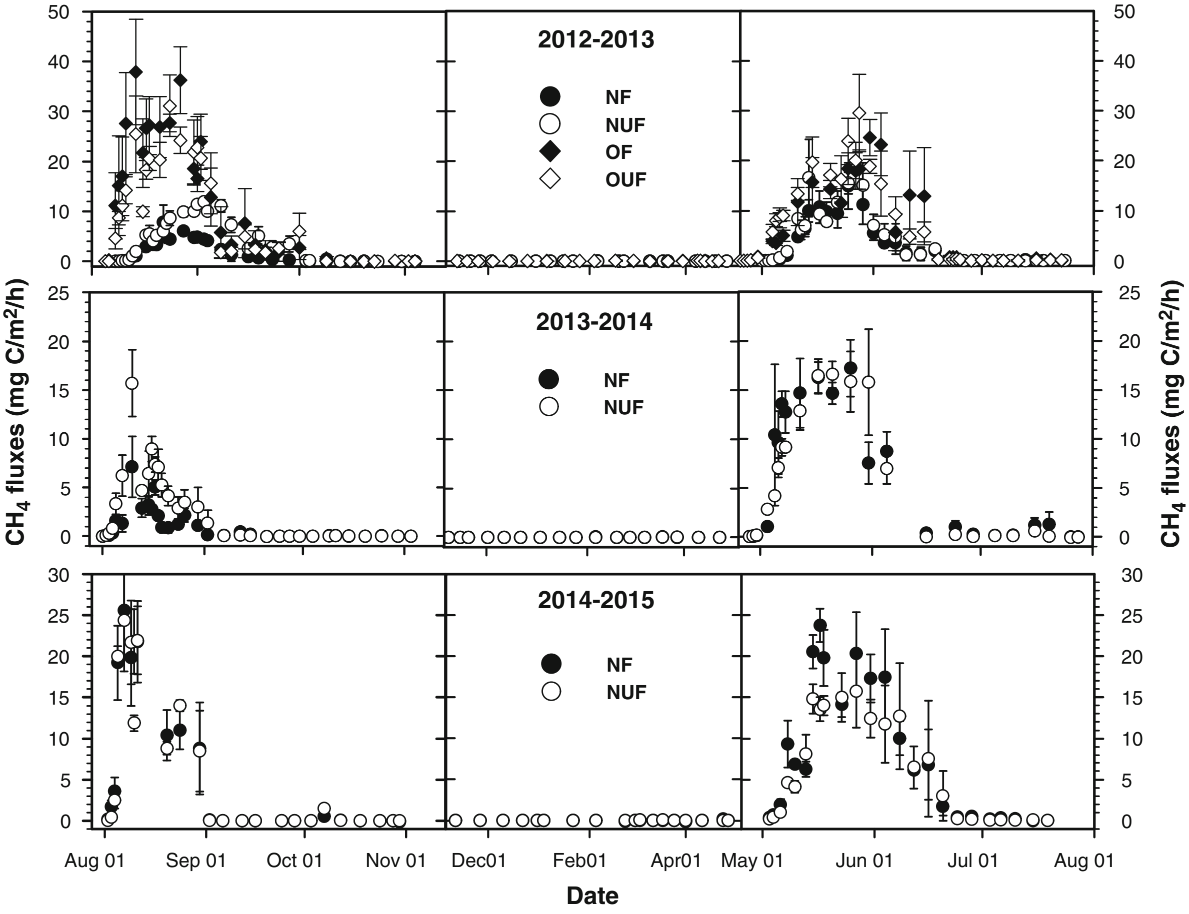

Fig. 2 Seasonal variations of CH_4 fluxes under the different treatments during the study years of 2012–2013 (*upper panel*), 2013–2014 (middle panel) and 2014–2015 (*lower panel*)

Table 2 Results of a two-way ANOVA for the main and interactive effects of land-use legacies and N fertilization on CH_4 fluxes

Source of variation	*df*	*SS*	*F*	*p*
Land-use legacies (years, L)	2	92.967	13.588	0.000
Fertilization (F)	1	0.073	0.032	0.860
L*F	2	2.845	0.416	0.744
Model	5	95.903	6.007	0.001
Error	14	45.612		

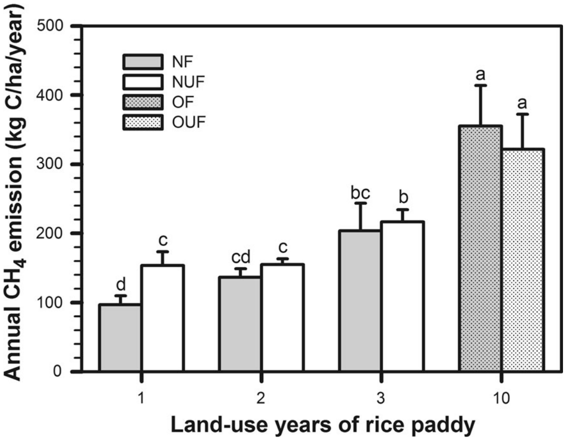

Fig. 3 Annual cumulative emissions of CH_4 in the different rice paddy land-use years. *Vertical bars* indicate standard errors of four replicates, and *different letters* indicate significant differences at the $p < 0.05$ level

lack of labile organic substrate in the upland soils (Hatala et al. 2012).

In the current study, the soil organic carbon (SOC) content in the new rice plots was significantly lower than in the old rice plots regardless of N fertilization management (Table 1). The long-term continuous cultivation of paddy rice favors SOC sequestration (Huang et al. 2012). The cropping duration of paddy rice could be the main reason for the significant difference of SOC between the new and old rice plots. Since SOC is an important substrate for CH_4 production (Watanabe et al. 1999), the significant differences of CH_4 emissions between the new and old rice plots were probably caused by lower SOC contents in the new rice plots inherited from upland plantations compared with the old rice plots.

Furthermore, CH_4 emissions significantly increased with time after establishment of the new rice fields (Fig. 3). The increasing trend of CH_4 emissions also reflected the effects of land-use legacies on CH_4 emissions and indicated that the effects of land-use legacies weakened with increasing time of new rice paddy cultivation (Eusufzai et al. 2010; Hatala et al. 2012; Nishimura et al. 2011). However, significant increases of SOC content in rice paddies usually takes decades (Zhang and He 2004), and during the study period, we found no significant increase of SOC content with land-use years of the new rice fields (Table 1). Given that climatic conditions were consistent and rice biomass did not significantly increase during the study period (Table 1), the increasing trend of annual CH_4 emissions in the new rice plots might be caused by other factors of land-use legacy, such as soil Eh.

Soil pH is an index that reflects proton concentration in the soil, which can influence redox reactions. The increase of soil pH in wetlands indicates a significant reduction of soil Eh, especially when strong oxidants (e.g., Fe^{3+}, Mn^{4+}) are present (Reddy and Delaune 2008). In the current study, soil pH significantly increased with the land-use years of the rice fields, which was consistent with CH_4 emissions

Table 3 Annual mean CH_4 fluxes (mg C/m^2/h, mean ± SE) and the NFE on CH_4 fluxes in rice plots under different land-use years

Land-use years of rice paddy	CH_4 fluxes		NFE
	Plots with fertilization	Plots without fertilization	
1	1.73 ± 0.17	2.61 ± 0.28	−0.87*a
2	2.24 ± 0.21	2.53 ± 0.07	−0.29b
3	4.00 ± 0.48	4.06 ± 0.30	−0.06b
10	6.69 ± 0.84	6.05 ± 0.73	0.63c

* The effect of N fertilization on CH_4 fluxes was significant at the 0.05 level

Different letters in the last column indicate that the NFE on CH_4 fluxes in the rice plots under different land-use years is significantly different at the $p < 0.05$ level

(Table 1). The increasing trend of soil pH with increasing time of the new rice plots indicates that soil Eh had decreased. A lower soil Eh could favor CH_4 production (Le Mer and Roger 2001), and thus, CH_4 emissions increased with the land-use years of the rice fields, which was consistent with that we found in the current study. CH_4 production in flooded soils is also very sensitive to pH (Yan et al. 2005), which has been reported to have an optimum range of 6.7–7.1 (Aulakh et al. 2001). However, Yan et al. (2005) reported that CH_4 emissions were much higher in soils with a pH of 5.0–5.5 compared to other soils. These findings suggest that soil pH per se may have little effect on CH_4 emissions in the current study.

Unfortunately, we did not measure the variation of soil Eh, CH_4 production and oxidation potential directly. To elucidate the effects of land-use legacies on CH_4 emissions in the new rice fields, further studies need to be conducted even though several studies have revealed the effects of land-use legacies on soil C and N cycles (McGrath et al. 2001; Zhang and He 2004; Eusufzai et al. 2010; Nishimura et al. 2011). However, our study may provide new clues for understanding CH_4 emissions from new double-rice paddies in subtropical regions (Yang et al. 2010; Dong et al. 2011; Shang et al. 2011; Banger et al. 2012). Our findings also suggest that regional GHG inventories should consider not only land use types but also land use history because of the effects of land-use legacies.

The N fertilization effect on CH_4 emissions

In the study, N fertilization significantly reduced CH_4 emissions in the first year after the establishment of the new rice plots (Table 3). Generally, CH_4 emissions from rice fields can be determined by the balance between CH_4 production and oxidation (Bodelier and Laanbroek 2004). Krüger et al. (2001) reported that 10–90% of produced CH_4 was oxidized in the rhizosphere of rice plants, owing to oxygen diffusion via rice aerenchyma in the rhizosphere (Wassmann and Aulakh 2000). Fertilization-enhanced plant growth can increase the oxygen supply to the rhizosphere through a better developed aerenchyma system, thus stimulating CH_4 oxidation (Aulakh et al. 2000). In the present study, N fertilization significantly increased rice biomass by approximately 66–110% in the new rice plots (Table 1), and previous studies have shown that greater rice biomass is highly associated with aerenchyma tissues (Aulakh et al. 2000). This suggests that fertilization could have increased CH_4 oxidation in the current study. Furthermore, better plant growth, as indicated by greater biomass, may also provide more organic substrates for methanogens through litter decomposition and root exudation (Jia et al. 2001), resulting in greater CH_4 production (Aulakh et al. 2001; Watanabe et al. 1999). When N fertilization-induced CH_4 oxidation exceeds its production (Xu and Inubushi 2004), N fertilization decreases the overall CH_4 emissions, as measured by CH_4 efflux in the current study. Otherwise, N fertilization may increase or have no effect on CH_4 emissions.

However, N fertilization had no significant effects on CH_4 emissions in the old rice plots and the new rice plots during the second and third year of cultivation even though N fertilization obviously increased rice biomass (Table 1). Moreover, N fertilization-induced CH_4 emissions increased with the land-use years of the rice paddy. Theoretically, the effects of N fertilization on CH_4 emissions could be influenced by site-specific environmental factors (Dong et al. 2011; Banger et al. 2012), which can lead to large interannual variability. In the study, N fertilization and water management practices were consistent between the old and new rice plots and among different years. Therefore, we examined soil temperature and pH, which could influence CH_4 emissions, as suggested by former studies (Lu et al. 2000; Krüger et al. 2001; Conrad 2002; Yan et al. 2005). During the study period, the patterns and variations of soil temperature were similar among the three years (Fig. 1), suggesting that temperature was unlikely to cause the changing effects of N fertilization on annual CH_4 emissions. Meanwhile, the changing trend of soil pH with land-use years was synchronous between the rice plots with and without fertilization (Table 1), suggesting that soil pH was also not the cause for the weakening negative effects of N fertilization. The results show that soil temperature and pH had little contribution to the variation of NFE in rice paddies, which are in line with a previous study (Banger et al. 2012).

We found that N fertilization-induced increase of soil NO_3^-–N decreased with land-use year in the new rice plots (Fig. 4). The increase of soil NO_3^-–N content induced by N fertilization was much larger in the 1-year rice plots than in the older ones, which might be the results of varying soil microbial

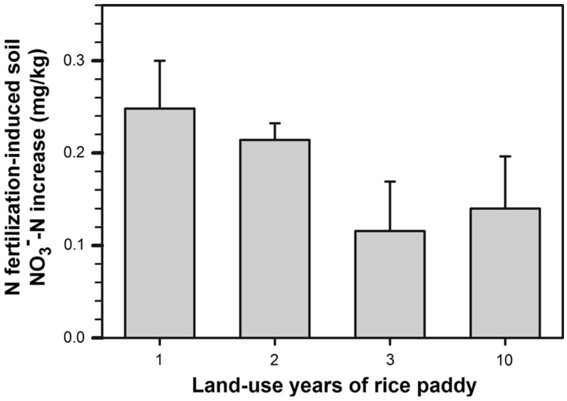

Fig. 4 N fertilization-induced soil NO_3^-–N increase in rice paddy soils with different land-use years. *Vertical bars* indicate the standard errors of four replicates

communities with the land-use years of the new rice plots. Chu et al. (2009) reported that soil ammonia-oxidizing bacteria (AOB) communities had larger population sizes and were more diverse in rice paddies recently converted from upland cultivation compared to consecutive rice paddies. The results suggested that relatively more NH_4^+–N from N fertilization was converted to NO_3^-–N in the first year of cultivation of the new rice paddy compared to the following two years and the old rice paddy. According to the literature, NO_3^-–N can reduce CH_4 emissions through competing for H_2 with denitrifying bacteria (Klüber and Conrad, 1998) or by inhibiting methanogenesis with toxic denitrification intermediates (Roy and Conrad, 1999). Therefore, the relatively stronger reduction effects of N fertilization on CH_4 emissions during the first year of cultivation of the new rice plots could be induced by larger differences in the soil NO_3^-–N concentration between the NF and NUF plots due to land-use legacies.

The results indicate that the effects of N fertilization on CH_4 emissions have been coupled with the effects of land-use legacies, as suggested by Perring et al. (2016), and that land-use legacies can interact with environmental changes such as N addition as well as modulate the carbon cycle. Considering that most studies were carried out in long-term rice paddies, our study may provide new clues for understanding the N fertilization effect on CH_4 emissions. However, the coupled effects of land-use legacies and environmental change are complicated (Perring et al. 2016). More studies need to be completed to understand the transforming states of the soil biotic and abiotic processes in the new rice paddies and their interaction with N fertilization.

Conclusions

Field results showed that CH_4 emissions from the new rice plots were significantly lower than the old rice plots. There was an increasing trend of annual CH_4 emissions with land-use years of cultivation in the new rice paddies. Annual CH_4 emissions increased with land-use year of the rice paddies, following the order of 1-year $<$ 2-year $<$ 3-year $<$ 10-year. N fertilization significantly reduced CH_4 emissions by 36.9% in the 1-year-old new rice plots, whereas it had no significant effect on CH_4 emissions in the older rice plots. These results suggest that land-use legacies can significantly affect CH_4 emissions in subtropical regions and may influence the N fertilization effect on CH_4 emissions. Land-use legacies and their possible coupled effect with N fertilization on CH_4 emissions should be considered in regional GHG inventories.

Acknowledgements This work was financially supported by the National Basic Research Program of China (973 Program, 2012CB417103) and the Forestry Department of Sichuan Province (Carbon Accounting Project, 2009-204). We thank the editors and anonymous reviewers for their constructive comments and suggestions in revising the manuscript. We thank Springer Nature English Language Editing for the language editing. We also thank the following persons for their help in field and laboratory work: Miaomiao Zhao, Jingdong Zou, Shihuang Zhang, Yubo Bin, Jiaying Lu, Ying Zhou, Yuanfen Huang and Ronghua Ou-yang.

References

Aulakh MS, Bodenbender J, Wassmann R, Rennenberg H (2000) Methane transport capacity of rice plants. II. Variations among different rice cultivars and relationship with morphological characteristics. Nutr Cycl Agroecosyst 58:367–375

Aulakh MS, Wassmann R, Bueno C, Rennenberg H (2001) Impact of root exudates of different cultivars and plant development stages of rice (*Oryza sativa* L.) on methane production in a paddy soil. Plant Soil 230:77–86

Banger K, Tian H, Lu C (2012) Do nitrogen fertilizers stimulate or inhibit methane emissions from rice fields? Glob Change Biol 18:3259–3267

Bodelier PLE, Laanbroek HJ (2004) Nitrogen as a regulatory factor of methane oxidation in soils and sediments. FEMS Microbiol Ecol 47:265–277

Bodelier PLE, Hahn AP, Arth IR, Frenzel P (2000a) Effects of ammonium-based fertilisation on microbial processes involved in methane emission from soils planted with rice. Biogeochemistry 51:225–257

Bodelier PLE, Roslev P, Henckel T, Frenzel P (2000b) Stimulation by ammonium-based fertilizers of methane oxidation in soil around rice roots. Nature 403:421–424

Bouwman AF (1996) Direct emission of nitrous oxide from agricultural soils. Nutr Cycl Agroecosyst 46:53–70

Cai Z, Mosier AR (2000) Effect of NH_4Cl addition on methane oxidation by paddy soils. Soil Biol Biochem 32:1537–1545

Cassman KG, Peng S, Olk DC, Ladha JK, Reichardt W, Dobermann A, Singh U (1998) Opportunities for increased nitrogen-use efficiency from improved resource management in irrigated rice systems. Field Crop Res 56:7–39

Chu H, Morimoto S, Fujii T, Yagi K, Nishimura S (2009) Soil ammonia-oxidizing bacterial communities in paddy rice fields as affected by upland conversion history. Soil Sci Soc Am Proc 73:2026–2031

Conrad R (2002) Control of microbial methane production in wetland rice field. Nutr Cycl Agroecosyst 64:59–69

Dong H, Yao Z, Zheng X, Mei B, Xie B, Wang R, Deng J, Cui F, Zhu J (2011) Effect of ammonium-based, non-sulfate fertilizers on CH_4 emissions from a paddy field with a typical Chinese water management regime. Atmos Environ 45:1095–1101

Eusufzai MK, Tokida T, Okada M, Sugiyama S, Liu GC, Nakajima M, Sameshima R (2010) Methane emission from rice fields as affected by land use change. Agric Ecosyst Environ 139:742–748

Feng JF, Chen CQ, Zhang Y, Song ZW, Deng AX, Zheng CY, Zhang WJ (2013) Impacts of cropping practices on yield-scaled greenhouse gas emissions from rice fields in China: a meta-analysis. Agric Ecosyst Environ 164:220–228

Hatala JA, Detto M, Sonnentag O, Deverel SJ, Verfaillie J, Baldocchi DD (2012) Greenhouse gas (CO_2, CH_4, H_2O) fluxes from drained and flooded agricultural peatlands in the Sacramento-San Joaquin Delta. Agric Ecosyst Environ 150:1–18

Huang S, Sun YN, Zhang WJ (2012) Changes in soil organic carbon stocks as affected by cropping systems and cropping duration in China's paddy fields: a meta-analysis. Clim Change 112:847–858

Jia Z, Cai Z, Xu H, Li X (2001) Effect of rice plants on CH_4 production, transport, oxidation and emission in rice paddy soil. Plant Soil 230:211–221

Klüber HD, Conrad R (1998) Effects of nitrate, nitrite, NO and N_2O on methanogenesis and other redox processes in anoxic rice field soil. FEMS Microbiol Ecol 25:301–318

Krüger M, Frenzel P, Conrad R (2001) Microbial processes influencing methane emission from rice fields. Glob Change Biol 7:49–63

Le Mer J, Roger P (2001) Production, oxidation, emission and consumption of methane by soils: a review. Eur J Soil Biol 37:25–30

Lindau CW, Bollich PK, Delaune RD, Patrick WH, Law J (1991) Effect of urea fertilizer and environmental factors on CH_4 emissions from a Louisiana, USA rice field. Plant Soil 136:195–203

Linquist BA, Adviento-Borbe MA, Pittelkow CM, van Kessel C, van Groenigen KJ (2012) Fertilizer management practices and greenhouse gas emissions from rice systems: a quantitative review and analysis. Field Crop Res 135:10–21

Lu WF, Chen W, Duan BW, Guo WM, Lu Y, Lantin RS, Wassmann R, Neue HU (2000) Methane emissions and mitigation options in irrigated rice fields in southeast China. Nutr Cycl Agroecosyst 58:65–73

McGrath DA, Smith CK, Gholz HL, Oliveira FA (2001) Effects of land-use change on soil nutrient dynamics in amazônia. Ecosystems 4:625–645

Myhre G, Shindell D, Bréon F-M et al (2013) Anthropogenic and Natural Radiative Forcing. In: Stocker TF, Qin D, Plattner G-K et al (eds) Climate change 2013: the physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change. Cambridge University Press, Cambridge, pp 659–740

Nishimura S, Akiyama H, Sudo S, Fumoto T, Cheng W, Yagi K (2011) Combined emission of CH_4 and N_2O from a paddy field was reduced by preceding upland crop cultivation. Soil Sci Plant Nutr 57:167–178

Perring MP, De Frenne P, Baeten L, Maes SL, Depauw L, Blondeel H, Carón MM, Verheyen K (2016) Global environmental change effects on ecosystems: the importance of land-use legacies. Glob Change Biol 22:1361–1371

Pittelkow CM, Adviento-Borbe MA, van Kessel C, Hill JE, Linquist BA (2014) Optimizing rice yields while minimizing yield-scaled global warming potential. Glob Change Biol 20:1382–1393

Reddy R, Delaune RD (2008) Beogeochemistry of wetland: science and applications. CRC Press, Taylor & Francis Group, Boca Raton

Roy R, Conrad R (1999) Effects of methanogenic precursors (acetate, hydrogen, propionate) on the suppression of methane production by nitrate in anoxic rice field soil. FEMS Microbiol Ecol 28:49–61

Sass RL, Fisher FM, Wang YB, Turner T, Jund MF (1992) CH_4 emission from rice fields-the effect of floodwater management. Glob Biogeochem Cycles 6:249–262

Schimel J (2000) Rice, microbes and methane. Nature 403:375–377

Schulze ED, Luyssaert S, Ciais P et al (2009) Importance of methane and nitrous oxide for Europe's terrestrial greenhouse-gas balance. Nat Geosci 2:842–850

Shang Q, Yang X, Gao C, Wu P, Liu J, Xu Y, Shen Q, Zou J, Guo S (2011) Net annual global warming potential and greenhouse gas intensity in Chinese double rice-cropping systems: a 3-year field measurement in long-term fertilizer experiments. Glob Change Biol 17:2196–2210

Smith P, Martino D, Cai Z et al (2007) Agriculture. In: Metz B, Davidson OR, Bosch PR, Dave R, Meyer LA (eds) Climate change 2007: mitigation. Contribution of working group III to the fourth assessment report of the intergovernmental panel on climate change. Cambridge University Press, Cambridge, pp 497–540

Smith P, Bustamante M, Ahammad H et al (2014) Agriculture, forestry and other land use (AFOLU). In: Edenhofer O, Pichs-Madruga R, Sokona Y, Minx CJ (eds) Climate change 2014: mitigation of climate change. Contribution of working group III to the fifth assessment report of the intergovernmental panel on climate change. Cambridge University Press, Cambridge, pp 811–922

Tang HM, Xiao XP, Tang WG, Wang K, Sun JM (2014) Effects of winter cover crops straws incorporation on CH_4 and N_2O emission from double-cropping paddy fields in southern China. PLoS ONE 9(10):e108322. doi:10.1371/journal.pone.0108322

Wang J, Zhang X, Xiong Z, Khalil MAK, Zhao X, Xie Y, Xing G (2012) Methane emissions from a rice agroecosystem in South China: effects of water regime, straw incorporation and nitrogen fertilizer. Nutr Cycl Agroecosyst 93:103–122

Wassmann R, Aulakh MS (2000) The role of rice plants in regulating mechanisms of methane emissions. Biol Fertil Soils 31:20–29

Watanabe A, Takeda T, Kimura M (1999) Evaluation of origins of CH_4 carbon emitted from rice paddies. J Geogr Res 104(19):23623–23629

Weller S, Janz B, Jörg L, Kraus D, Racela HSU, Wassmann R, Butterbach-Bahl K, Kiese R (2016) Greenhouse gas emissions and global warming potential of traditional and diversified tropical rice rotation systems. Glob Change Biol 22:432–448

Xie B, Zheng X, Zhou Z, Gu J, Zhu B, Chen X, Shi Y, Wang Y, Zhao Z, Liu C, Yao Z, Zhu J (2010) Effects of nitrogen fertilizer on CH_4 emission from rice fields: multi-site field observations. Plant Soil 326:393–401

Xu X, Inubushi K (2004) Effects of N sources and methane concentrations on methane uptake potential of a typical coniferous forest and its adjacent orchard soil. Biol Fertil Soils 40:215–221

Yagi K, Tsuruta H, Kanda K, Minami K (1996) Effect of water management on methane emission from a Japanese rice paddy field: automated methane monitoring. Glob Biogeochem Cycles 10:255–267

Yan X, Yagi K, Akiyama H, Akimoto H (2005) Statistical analysis of the major variables controlling methane emission from rice fields. Glob Change Biol 11:1131–1141

Yang X, Shang Q, Wu P, Liu J, Shen Q, Guo S, Xiong Z (2010) Methane emissions from double rice agriculture under long-term fertilizing systems in Hunan, China. Agric Ecosyst Environ 137:308–316

Zhang M, He Z (2004) Long-term changes in organic carbon and nutrients of an Ultisol under rice cropping in southeast China. Geoderma 118:167–179

Zou J, Huang Y, Jiang J, Zheng X, Sass RL (2005) A 3-year field measurement of methane and nitrous oxide emissions from rice paddies in China: effects of water regime, crop residue, and fertilizer application. Glob Biogeochem Cycl 19:GB2021