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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

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Abstract

The impact of agricultural management on global warming potential (GWP) and greenhouse gas intensity (GHGI) is not well documented. A long-term fertilizer experiment in Chinese double rice-cropping systems initiated in 1990 was used in this study to gain an insight into a complete greenhouse gas accounting of GWP and GHGI. The six fertilizer treatments included inorganic fertilizer [nitrogen and phosphorus fertilizer (NP), nitrogen and potassium fertilizer (NK), and balanced inorganic fertilizer (NPK)], combined inorganic/organic fertilizers at full and reduced rate (FOM and ROM), and no fertilizer application as a control. Methane (CH_4) and nitrous oxide (N_2O) fluxes were measured using static chamber method from November 2006 through October 2009, and the net ecosystem carbon balance was estimated by the changes in topsoil (0–20 cm) organic carbon (SOC) density over the 10-year period 1999–2009. Long-term fertilizer application significantly increased grain yields, except for no difference between the NK and control plots. Annual topsoil SOC sequestration rate was estimated to be $0.96\,\mathrm{t\,C\,ha^{-1}\,yr^{-1}}$ for the control and 1.01–$1.43\,\mathrm{t\,C\,ha^{-1}\,yr^{-1}}$ for the fertilizer plots. Long-term inorganic fertilizer application tended to increase CH_4 emissions during the flooded rice season and significantly increased N_2O emissions from drained soils during the nonrice season. Annual mean CH_4 emissions ranged from $621\,\mathrm{kg\,CH_4\,ha^{-1}}$ for the control to $1175\,\mathrm{kg\,CH_4\,ha^{-1}}$ for the FOM plots, 63–83% of which derived from the late-rice season. Annual N_2O emission averaged 1.15–4.11 $\mathrm{kg\,N_2O\text{–}N\,ha^{-1}}$ in the double rice-cropping systems. Compared with the control, inorganic fertilizer application slightly increased the net annual GWPs, while they were remarkably increased by combined inorganic/organic fertilizer application. The GHGI was lowest for the NP and NPK plots and highest for the FOM and ROM plots. The results of this study suggest that agricultural economic viability and GHGs mitigation can be simultaneously achieved by balanced fertilizer application.

Keywords: carbon dioxide, GHGI, GWP, long-term fertilizer experiment, methane, nitrous oxide, rice paddy, soil organic carbon

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Introduction

Atmospheric carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are most potent long-lived greenhouse gases (GHGs) that contribute to global warming. Increasing concentrations of CO_2, CH_4 and N_2O in 2005 have led to a combined radiative forcing of $2.30\,\mathrm{W\,m^{-2}}$, consisting of $1.66\,\mathrm{W\,m^{-2}}$ by CO_2, $0.48\,\mathrm{W\,m^{-2}}$ by CH_4 and $0.16\,\mathrm{W\,m^{-2}}$ by N_2O (Forster *et al*., 2007). Agriculture accounted for an estimated emission of 5.1–6.1 Pg CO_2-equivalents $\mathrm{yr^{-1}}$, contributing 10–12% to the total global anthropogenic emissions of GHGs in 2005 (Smith *et al*., 2007). While agriculture releases significant amount of CH_4 and N_2O to the atmosphere, the net emission of CO_2 equivalents from farming activities can potentially be decreased by changing agricultural management to increase soil organic matter content and/or decrease CH_4 and N_2O emissions (Kroeze *et al*., 1999; Robertson *et al*., 2000; Follett, 2001; Mosier *et al*., 2006; Smith *et al*., 2008). The global technical mitigation potential from agriculture (excluding fossil fuel offsets from biomass) is estimated to be approximately 5.5–6.0 Pg CO_2-equivalents $\mathrm{yr^{-1}}$ by 2030 (Smith *et al*., 2008).

Many agricultural practices have been advocated to mitigate GHGs emission, generally through the mechanisms of enhancing removals from atmosphere (sequestrating), reducing emissions, and avoiding (or displacing) emissions (Smith *et al*., 2008). Converting atmospheric CO_2 into stable organic carbon pools in the soil can sequester CO_2, while changes in soil carbon storage depend on the balance between carbon input

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and output. Thus, optimizing agricultural managements can increase soil carbon by increasing carbon input and/or decreasing carbon output. Available field experiment results have widely proved that fertilizer application and straw return can increase soil carbon and sequester carbon from the atmosphere (e.g. Cole *et al*., 1993; Rasmussen & Parton, 1994; Dumanski *et al*., 1998; Halvorson *et al*., 1999; Lu *et al*., 2009). CH_4 is produced when organic materials decompose in oxygen-deprived conditions, such as rice grown under flooded conditions (Mosier *et al*., 1998; Sass *et al*., 1999). Midseason drainage has been proved to be an effective option for mitigating CH_4 emissions (typically reduced by 35–70%) from rice paddies (Cai, 1997; Mishra *et al*., 1997; Zou *et al*., 2005). N_2O is generated primarily by the microbial processes of nitrification and denitrification in soils and manures. Balanced fertilizer management can increase fertilizer-use efficiency and fairly meets plant requirements, and thus practices that deliver added N more efficiently to crops often suppress N_2O emission (Bouwman, 2001).

Although several agricultural practices are available to targeting a specific GHG (CH_4 or N_2O) mitigation or soil organic carbon (SOC) sequestration, agricultural GHG fluxes are complex, such as trade-offs among them. Balanced fertilizer application can raise crop yield as well as biomass, which will increase soil carbon sequestration due to biomass input into soil from crop residues and roots, but it may be off-set by N_2O emissions increased with fertilizer (Six *et al*., 2004). Crop residue amendment can increase SOC sequestration, while it often causes substantial CH_4 emission from rice paddies (Adhya *et al*., 2000; Cai *et al*., 2000; Zou *et al*., 2005; Ma *et al*., 2009). Shifting from continuous waterlogging to midseason drainage leads to a drop in CH_4 flux but an increase in N_2O flux in rice paddies (Cai *et al*., 1997; Zheng *et al*., 2000; Zou *et al*., 2005). Consequently, the overall balance between the net exchange of these gases constitutes the net global warming potential (GWP) of a crop production system (Robertson & Grace, 2004). Taking effective agriculture management strategy for mitigating climatic impacts requires a complete perspective on the agriculture impacts on radiative forcing (Frolking *et al*., 2004; Robertson & Grace, 2004; Mosier *et al*., 2006).

The concept of GWP, one type of simplified index based upon radiative properties, was introduced in order to estimate the potential future impacts of emissions of different gases upon the climate system in a relative sense (Lashof & Ahuja, 1990). In GWP estimation, CO_2 is typically taken as the reference gas, and an increase or reduction in emission of CH_4 and N_2O is converted into 'CO_2-equivalents' by means of their GWPs. A positive GWP number represents a net source of CO_2 equivalents and a negative value indicates net sinks of atmospheric GHG. Another concept, greenhouse gas intensity (GHGI), was introduced to relate agricultural practices to GWP (Li *et al*., 2006; Mosier *et al*., 2006; Qin *et al*., 2010). This term is calculated by dividing GWP by crop yield, regarding climatic impacts of agriculture in terms of per kg of yield. To our knowledge, little is known about the impacts of agricultural management on GHGI in typical farming systems (Li *et al*., 2006; Mosier *et al*., 2006; Qin *et al*., 2010).

Rice is the staple food for nearly 50% of the world's people, mainly in Asia. China is the most important rice producing country in the world, and its planting area accounts for about 20% of the world total and occurs on 23% of all cultivated land in China (Frolking *et al*., 2002). A great amount of field measurements and model studies have focused on GHGs in rice paddies, dedicating to an overall estimate of GHGs in Chinese rice paddies (Cai *et al*., 1997; Zheng *et al*., 2000; Li *et al*., 2002, 2004; Zou *et al*., 2005). The SOC sequestration of paddy soils ranged from 1.3 to 5.1 Pg, dependent on soil profile depth and estimate methods, and the observed SOC sequestration rate of the paddy topsoils ranged from 0.13 to 2.20 $t\,C\,ha^{-1}\,yr^{-1}$, with a weighted mean of 0.37–0.40 $t\,C\,ha^{-1}\,yr^{-1}$ (Pan *et al*., 2003; Huang & Sun, 2006; Liu *et al*., 2006; Xie *et al*., 2007; Liao *et al*., 2009; Sun *et al*., 2009). The total CH_4 emissions from Chinese rice paddies were estimated to be 6–10 $Tg\,yr^{-1}$ in the 1990s (Huang *et al*., 2004a; Yan *et al*., 2009). Nitrogen input has increased from 87.5 $kg\,N\,ha^{-1}$ in the 1950s to 224.6 $kg\,N\,ha^{-1}$ in the 1990s, accompanied by direct N_2O emissions increased from 9.6 to 32.3 Gg N_2O–N in Chinese paddy rice production over the period 1950s–1990s (Zou *et al*., 2009a). While these studies provided an insight into GHGs inventory in Chinese rice paddies, we know little about the accounting of net annual GWP and GHGI in rice paddies in China since simultaneous measurements of CO_2, CH_4 and N_2O emissions have been rarely taken over the whole annual cycle (Zou *et al*., 2004). To our knowledge, only the DNDC model studies have projected the impacts of agricultural management on the net GWPs and GHGIs in China's rice paddies, but they are highly needed to be confirmed by field measurements (Li *et al*., 2005, 2006).

A long-term fertilizer experiment initiated in 1990 was used to evaluate the impacts of fertilizer application regime on the net annual GWPs and GHGIs in double rice-cropping systems in Hunan province China, where is dominated by double rice-cropping systems. Annual double rice-cropping (15%) and double rice-other crop (41%) rotations represent the typical rice production regime, together accounting for 56% of rice planting area in China (Frolking *et al*., 2002). We present field measurements of CH_4 and N_2O emissions from double rice-cropping systems under various long-term fertilizer management regimes over the three annual rotation cycles of the 2006 winter season to the 2009 late-rice growing season. The net ecosystem carbon balance was estimated by the changes in topsoil organic carbon density (SOCD) over the period 1999–2009. To draw a conclusion by integrating SOC, CH_4 and N_2O measurements over the three annual rotations, a minor part of the CH_4 flux data in the 2007 double rice-growing seasons having been previously reported in Yang *et al.* (2010) was here used again. The objectives of this study are to gain an insight into a complete GHG accounting of GWP and GHGI as affected by long-term fertilizer application in double rice-cropping systems, and thereby to optimize agricultural management strategies for simultaneously achieving grain yields and mitigating climatic impacts of double rice production in China.

Materials and methods

Experiment site

A 3-year field experiment began in the 2006 winter season was carried out in a long-term fertilizer experimental grid at Taoyuan Agro-ecological Experimental Station. It is located at the bottom of slope in a typical hilly agricultural area in Hunan Province, China (28°55′N, 111°30′E; altitude: 92.2–125.3 m), where cropping regime is dominated by the double rice-cropping systems. The paddy soil is classified as stagnic anthrosols developed from Quaternary red clay. The region is characterized by the subtropical humid monsoon climate, with an annual average air temperature of 16.5 °C, precipitation of 1448 mm, sunshine of 15:13 h, and frost-free period of 283 days. The monthly climate information and the soil physicochemical properties and nutrients content during the experimental period are shown in Appendices S1 and S2, respectively.

Fertilization treatments

The long-term fertilizer experimental grid has been established since 1990. In this long-term fertilizer experiment, a randomized block experiment with three replicates was established, with six different fertilizer treatments. Each field plot is 4.1 m × 8.1 m. The six fertilizer treatments represent imbalanced/balanced inorganic fertilizer or combined inorganic/organic fertilizer regimes that are currently adopted at local double rice-cropping production (Appendix S3). Specifically, the six fertilizer treatments included nutrient potassium deficit (NP), nutrient phosphorus deficit (NK), balanced mineral nutrients (NPK), combined inorganic/organic fertilizers at full rate (FOM) or at reduced rate (ROM), and no fertilizer application as a control (Control). In the two cropping cycles, the total amount of nutrients N, P, and K were comparable among the treatments except for the ROM treatment with reduced rate at two-thirds of N, two-thirds of P and one-thirds of K of other treatments, varying in splitting ratio within the two crops. Urea for N, calcium superphosphate for P and potassium chloride for K were used throughout the experiments. Urea was applied with two splits for the early-rice season, 40% as basal fertilizer and 60% as tillering fertilizer and three splits for the late-rice season, 40% as basal fertilizer, 50% as tillering fertilizer and 10% as panicle fertilizer. The P and K fertilizers were applied as basal fertilizers before rice transplanting. The basal fertilizer, applied 2 days before rice transplanting, was well incorporated into the soil by plowing to 10–20 cm depth, and the top-dressing was surface broadcasted.

The plots under Control, NP, NK and NPK treatments were fallowed in winter and all rice straws including the early and late rice were harvested and removed out of the field. In the FOM and ROM plots, green manure (Chinese milk vetch) grown in the winter season was harvested and *in situ* ploughed into the soil before the early-rice transplanting. The application rates of fresh Chinese milk vetch with the water content of 88–91% were shown in Appendix S3. In addition, the harvesting crop straw of early or late rice (10 cm pieces) was *in situ* incorporated at full rate at the depth of 10–15 cm just before late-rice transplanting or after harvesting for the FOM plots, respectively (Appendix S3). Similar to the FOM plots, crop straw was incorporated at half rate for the ROM plots. The nitrogen content of incorporated Chinese milk vetch and crop residue averaged 2.60% and 0.93%, respectively.

Cropping regime and water management

In 2007 and 2008, local rice cultivars (*Oryza sativa* L.), Xiang 24 and Jingyou 27, were used for the early and late rice-cropping seasons, respectively. In 2009, the early and late rice cultivars were Xiang 45 and Tyou 207, respectively. In 2007–2009, rice seedlings were transplanted on April 26–May 2 and harvested on July 11–18 for the early rice crop, followed by the late rice-cropping season with transplanting on July 16–21 and harvesting on October 10–17 (Appendix S1). The intervals between the two rice-cropping seasons were about 3–5 days. In the winter season, Chinese milk vetch was grown in rice paddies for the FOM and ROM plots, whereas fallow was kept for the other fertilizer treatment plots.

All field plots were drained in the winter season. Consistent with water management in local double rice-cropping systems, flooding was initiated 1 week before early-rice transplanting, and maintained until 7–10 days before rice harvesting during the early rice-cropping season. During the late rice-cropping season, the water regime is similar to that during the early rice-cropping season but one to two drainage episodes were implemented at the late tillering stage. The waterlogging depth was maintained at 5 cm depth in rice paddies with an irrigation-drainage system. In addition, pesticide and herbicide management followed the local practices.

TopSOC sequestration estimates

We collected soil samples from all experimental plots at the plough layer depth of 0–20 cm before the early-rice transplanting to measure topsoil organic carbon (SOC) content. Soil samples were collected in each year over the period 1999–

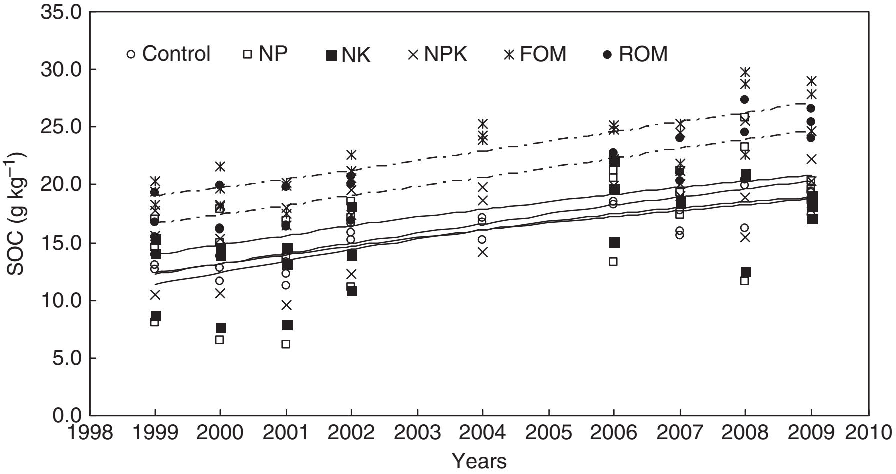

Fig. 1 Interannual changes in soil organic carbon (SOC) content in double-rice cropping systems under various fertilization regime treatments from 1999 to 2009. Data were fitted by the logistic regression model, simulated parameters and statistics see Table 1.

2009 except for 2003 and 2005. Each sample of about 1 kg was a composite of six subsamples randomly taken within a plot. Visible plant detritus and any fragments were removed after air-drying at room temperature, ground to pass a 2 mm sieve, and a portion subsequently ground in a porcelain mortar to pass a 0.15 mm sieve for SOC measurement. The SOC content of soil samples was determined with a CNS elemental analyzer (Variomax CNS Analyser, Elementar GmbH, Hanau, Germany) after decarbonization with HCl.

Over the period 1999–2009, topsoil organic carbon content (SOC, g C kg^{-1}) increased annually, but it tended to be leveling-off for the control and inorganic fertilizer treatments (Fig. 1). Although the SOC content tended to be linearly increased for the FOM and ROM treatments, the annual increase rates of SOC simulated by the linear and logistic regression models did not significantly differ (e.g. 0.80 vs. 0.81 g C kg yr^{-1} for the FOM). Therefore, we adopted logistic regression model to simulate annual increase rate of SOC content for all treatments [Eqn (1), Table 1]:

$$SOC = K_{\mathrm{SOC}}/(1 + be^{-rt}), \tag{1}$$

where K_{SOC} and r refer to the saturation content and relative increase rate of SOC, respectively. b is the model parameter and t is the duration of the experiment (years since 1999).

Annual topsoil organic carbon sequestration rate (*SOCSR*, t C ha^{-1} yr^{-1}) was estimated on the basis of topsoil SOC content increase rate (d_{SOC}/d_t, g C kg^{-1} yr^{-1}). Based on the Eqn (1), the d_{SOC}/d_t was estimated by the following equation [Eqn (2), Table 1]:

$$d_{\mathrm{SOC}}/d_t = rSOC_t(1 - SOC/K_{\mathrm{SOC}}) \tag{2}$$

where SOC_t refers to the SOC content in the experimental years of 2007–2009. Thereafter, the *SOCSR* was estimated by using the following equation (Pan *et al.*, 2003; Huang & Sun, 2006; Liao *et al.*, 2009; Lu *et al.*, 2009):

$$SOCSR = \mathrm{d}_{\mathrm{SOC}}/d_t \times \gamma \times (1 - \delta_{2\,\mathrm{mm}}/100) \times 20/10, \tag{3}$$

Where γ and $\delta_{2\,\mathrm{mm}}$ are the average bulk density (g cm^{-3}) and the gravel content (>2 mm, 3–9%) of topsoil (0–20 cm) under a specific fertilizer treatment (Appendix S2). The numbers 20 and 10 in the equation are the topsoil depth and the area conversion coefficient, respectively.

CH_4 and N_2O measurements

The measurements of CH_4 and N_2O emissions were initiated in the 2006 winter season, and continued over the three annual cycles of 2006–2009. The CH_4 and N_2O fluxes were measured using a static closed chambers method. Sampling chamber was made of PVC with a size of 67 × 67 × 110 cm. It was equipped with two circulating fans inside to ensure sufficient gas mixing and wrapped with a layer of sponge and aluminum foil to minimize air temperature changes inside the chamber during the period of sampling. Over each rice growing season or nonrice growing season, a PVC frame was permanently fixed into a random site in each plot. The top edge of the frame had a groove for filling with water to seal the rim of the chamber. Gas samples were taken from 08:00 through 10:00 hours since the soil temperature during this period was close to the mean daily soil temperature (Zou *et al.*, 2005). Air samples from the chamber headspace were collected using 100 mL plastic syringes fitted with three-way stopcocks via a Teflon tube (with an outer diameter of 1/8 in) connected to the chamber. About 20 mL gas samples were then immediately transferred to pre-evacuated vials with rubber stoppers. The air temperature inside the chamber was monitored during gas collection. Over the three annual cycles, fluxes of CH_4 and N_2O were measured in triplicate plots for all treatments generally twice a week (every 3 or 4 days), except for about 10-day interval during the nonrice growing periods.

The mixing ratios of CH_4 and N_2O were simultaneously analyzed with a modified gas chromatograph (GC-14B, Shimadzu Scientific Instruments Inc., Kyoto, Japan) equipped with a flame ionization detector (FID) and an electron capture detector (ECD). The oven was operated at 55 °C, the ECD at 330 °C, and the FID at 200 °C, respectively. The carrier gas (N_2) flow rate was 30 mL min^{-1}. Fluxes were determined from the slope of mixing ratio change in four samples, taken at 0, 10, 20

Table 1 Parameters and statistics for the logistic regression model and the estimated relative increase rate of soil organic carbon (SOC) content (d_{SOC}/d_t) over the period 2007–2009

	Parameters			Statistics				d_{SOC}/dt			
Treatment	K_{soc}	b	r	R^2	RMSE	MEF	P	2007	2008	2009	Average
Control	22.07	0.91	0.15	0.72	1.46	0.70	<0.001	0.62	0.41	0.43	0.49
NP	26.43	1.32	0.13	0.35	4.27	0.30	<0.01	0.70	0.64	0.68	0.67
NK	21.10	1.04	0.20	0.48	3.10	0.43	<0.001	0.45	0.51	0.52	0.49
NPK	24.53	0.88	0.14	0.41	3.03	0.38	<0.001	0.52	0.53	0.50	0.52
FOM	47.23	1.60	0.07	0.70	1.89	0.69	<0.001	0.82	0.80	0.80	0.80
ROM	37.50	1.38	0.09	0.72	1.93	0.71	<0.001	0.82	0.78	0.74	0.78

Logistic model $SOC = K_{soc}/(1 + be^{-rt})$, K_{soc} and r refer to the saturation content and relative increase rate of SOC, respectively. b is the model parameter and t is the duration of the experiment (years since 1999). R^2, coefficient of determination; RMSE, root mean squared error; MEF, modeling efficiency. $d_{SOC}/d_t = rSOC_t[1-SOC_t/K_{soc})]$, SOC_t refers to the SOC content in experimental years of 2007–2009.

and 30 min after chamber closure. Sample sets were rejected unless they yielded a linear regression value of $r^2 > 0.90$. Average fluxes and standard deviations of CH_4 and N_2O were calculated from triplicate plots. Seasonal amounts of CH_4 and N_2O emissions were sequentially accumulated from the emissions between every two adjacent intervals of the measurements (Zou *et al.*, 2005, Tables 2 and 4).

Net GWPs and GHGI estimates

To understand a complete accounting of the climatic impact of double rice-cropping systems under different fertilizer regimes, we adopted the IPCC factors to calculate the combined GWPs for 100 years by using the following equation (Forster *et al.*, 2007):

$$GWP = 25 \times CH_4 + 298 \times N_2O - SOCSR \times 44/12 (\text{kg } CO_2\text{-equivalents ha}^{-1}\text{ yr}^{-1}). \quad (4)$$

Thereafter, the GHGI is calculated by dividing GWP by rice grain yield (Li *et al.*, 2006; Mosier *et al.*, 2006; Qin *et al.*, 2010):

$$GHGI = GWP/\text{grain yield } (\text{kg } CO_2\text{-equivalents kg}^{-1}\text{ grain yield yr}^{-1}). \quad (5)$$

Other data measurements

The field topsoil samples were collected from all experimental plots before the early-rice transplanting in 2007–2009 to measure soil pH and soil nutrients content (Appendix S2). Soil texture was measured in 2001. Soil bulk density was measured in 1999, 2001, 2004 and 2007, and thus the bulk density data shown in Appendix S2 represent the average value of 1999–2009. Soil properties analyses were directed by the Chinese Soil Society guidelines (Lu, 2000). Grain yields were measured at physiological maturity by hand harvesting two rows 2 m long per plot. Grain yields of the rice were determined at harvest by oven drying to a constant weight at approximately 70%.

Statistical analyses

Increases in SOC content with year were simulated by a logistic regression model [Eqn (1)]. The three statistics RMSE (root mean squared error), R^2 (coefficient of determination) and MEF (modeling efficiency) are used for model evaluation (Table 1). Differences in seasonal CH_4 and N_2O emissions from double rice-cropping systems over the period 2007–2009 as affected by fertilizer treatment, year and their interaction were examined by using a two-way analysis of variance (ANOVA, Table 3). Differences in seasonal CH_4 and N_2O emissions among treatments over 2007–2009 were further examined by the Tukey's multiple range test (Table 2). Statistical analyses were carried out using JMP, ver. 5.1 (SAS Institute, Cary, NC, USA, 2003).

Results

Topsoil organic carbon sequestration

Compared with the control, long-term fertilizer application significantly increased topsoil SOC content (Appendix S1, Fig. 1). Over the experimental period 2007–2009, mean SOC content (26.0 g C kg^{-1}) was highest for the FOM plots, which was 44% greater than that of the control plots. Relative to the control, SOC content was 12–32% greater for the plots under long-term inorganic fertilizer application (NP, NK and NPK). Over the 10-year period 1999–2009, all treatments exhibited a significant increase in SOC content (Fig. 1). Topsoil SOC content has increased from 13.2, 14.6 and 19.3 g C kg^{-1} in 1999 to 18.7, 20.4 and 27.1 g C kg^{-1} in 2009 for the control, NPK and FOM plots, respectively. The simulated relationship projected topsoil SOC content to be leveling-off at 21.1–22.1 g C kg^{-1} for the NK and control, 24.5–26.4 g C kg^{-1} for the NPK and NP and 37.5–47.2 g C kg^{-1} for the combined inorganic/organic fertilizer treatments (Table 1).

Table 2 Seasonal $CH_4$and N_2O emissions and rice grain yield from the different fertilizer treatments during the early- and late-rice growing seasons in 2007–2009

		Early-rice season			Late-rice season		
Year	Treatment	CH_4 ($kg\,ha^{-1}$)	N_2O ($kg\,N\,ha^{-1}$)	Yield ($t\,ha^{-1}$)	CH_4 ($kg\,ha^{-1}$)	N_2O ($kg\,N\,ha^{-1}$)	Yield ($t\,ha^{-1}$)
2007	Control	203.4 ± 55.9	0.04 ± 0.06	3.33 ± 0.96	455.6 ± 38.0	−0.03 ± 0.07	4.34 ± 0.83
	NP	213.5 ± 32.1	0.13 ± 0.06	5.62 ± 0.71	478.3 ± 235.9	0.21 ± 0.09	5.13 ± 0.68
	NK	210.2 ± 103.7	0.13 ± 0.13	4.21 ± 1.34	524.7 ± 177.7	0.26 ± 0.20	4.91 ± 1.13
	NPK	279.2 ± 54.3	0.00 ± 0.10	7.03 ± 1.04	549.1 ± 165.0	0.11 ± 0.18	6.28 ± 0.01
	FOM	350.7 ± 128.0	0.22 ± 0.01	8.81 ± 0.12	861.7 ± 151.9	0.52 ± 0.25	6.29 ± 0.19
	ROM	347.8 ± 107.2	0.05 ± 0.07	7.55 ± 0.35	696.5 ± 166.2	0.27 ± 0.04	6.06 ± 0.08
2008	Control	140.6 ± 12.8	−0.06 ± 0.10	3.01 ± 0.43	352.3 ± 60.7	0.11 ± 0.01	3.69 ± 0.23
	NP	149.8 ± 52.9	0.21 ± 0.14	5.31 ± 0.74	494.7 ± 142.6	0.31 ± 0.09	4.50 ± 0.25
	NK	152.6 ± 32.6	0.20 ± 0.03	2.63 ± 0.92	432.9 ± 146.1	0.14 ± 0.04	3.71 ± 1.16
	NPK	188.5 ± 43.3	0.13 ± 0.07	6.47 ± 0.61	533.9 ± 183.4	0.04 ± 0.20	5.17 ± 0.44
	FOM	272.3 ± 79.0	0.30 ± 0.03	7.05 ± 0.08	737.4 ± 145.9	0.29 ± 0.13	5.46 ± 0.32
	ROM	223.2 ± 78.1	0.27 ± 0.04	5.44 ± 0.26	712.1 ± 235.3	0.16 ± 0.19	5.41 ± 0.12
2009	Control	193.7 ± 10.2	0.14 ± 0.18	2.19 ± 0.51	515.8 ± 84.5	0.09 ± 0.05	2.97 ± 0.46
	NP	210.5 ± 39.4	0.11 ± 0.10	4.56 ± 0.59	500.0 ± 164.2	0.38 ± 0.08	4.70 ± 0.09
	NK	213.3 ± 66.1	0.21 ± 0.04	2.01 ± 0.76	409.7 ± 175.0	0.11 ± 0.17	2.80 ± 0.82
	NPK	293.4 ± 71.2	0.21 ± 0.42	5.27 ± 0.14	535.2 ± 70.8	0.30 ± 0.13	4.75 ± 0.20
	FOM	429.1 ± 49.3	0.10 ± 0.43	5.92 ± 0.22	881.8 ± 158.2	0.41 ± 0.08	4.99 ± 0.12
	ROM	412.3 ± 115.8	0.25 ± 0.02	5.59 ± 0.46	843.8 ± 179.3	0.22 ± 0.09	4.67 ± 0.22
2007–2009*	Control	179.2 ± 41.3c	0.04 ± 0.14a	2.84 ± 0.78d	441.2 ± 90.5c	0.06 ± 0.08c	3.67 ± 0.77c
	NP	191.3 ± 48.1b	0.15 ± 0.10a	5.16 ± 0.75c	491.0 ± 160.7c	0.30 ± 0.10ab	4.78 ± 0.46b
	NK	192.0 ± 70.2b	0.18 ± 0.08a	2.95 ± 1.33d	455.7 ± 153.8c	0.17 ± 0.15bc	3.81 ± 1.29c
	NPK	253.7 ± 70.0ab	0.11 ± 0.24a	6.26 ± 0.99b	539.4 ± 128.5bc	0.15 ± 0.19bc	5.40 ± 0.73ab
	FOM	350.7 ± 104.3a	0.20 ± 0.23a	7.26 ± 1.27a	827.0 ± 148.1a	0.41 ± 0.18a	5.58 ± 0.60a
	ROM	327.8 ± 121.2a	0.19 ± 0.11a	6.20 ± 1.07b	750.8 ± 183.5ab	0.22 ± 0.11abc	5.38 ± 0.62ab

*Mean ± SD, different letters within the same column indicate statistical differences in variables mean among treatments over the 2007–2009 seasons by the Tukey's multiple range test ($P<0.05$). Seasonal CH_4 emissions in 2007 were redrawn from Yang *et al.* (2010).

Based on topsoil SOC content and bulk density, we estimated topsoil SOC density (SOCD) to be 36.4–48.2 $t\,C\,ha^{-1}$ in 2009, 15–30% greater than in 1999. The simulated relationship predicted that SOCD increase rate ranged from 0.49 $g\,C\,kg^{-1}\,yr^{-1}$ for the control to 0.80 $g\,C\,kg^{-1}\,yr^{-1}$ for the FOM treatments over the period 2007–2009 (Table 1). Correspondingly, annual topsoil SOC sequestration rate was estimated to be 0.96 $t\,C\,ha^{-1}\,yr^{-1}$ for the control and 1.01–1.43 $t\,C\,ha^{-1}\,yr^{-1}$ for the fertilizer treatments, with an average of 1.21 $t\,C\,ha^{-1}\,yr^{-1}$ for the experimental double rice-cropping paddy plots over the period 2007–2009 (Table 5).

CH_4 emission

During the early-rice growing season under continuous flooding, CH_4 fluxes were gradually increased in the early stage and stepped down in the late stage (Fig. 2). The fluxes of CH_4 showed a slight peak in the mid-season for the plots with inorganic fertilizer applications (NP, NK and NPK) and the control, while combined inorganic/organic fertilizer applications induced two higher peaks for the FOM and ROM plots. Seasonal total of CH_4 emissions significantly varied with fertilizer treatment and year, but it was not significantly affected by their interaction in the early-rice growing season (Tables 2 and 3). Averaged CH_4 emission was significantly lower in the 2008 early-rice season than in the 2007 and 2009 seasons ($P<0.05$). Relative to the control, inorganic fertilizer application increased CH_4 emissions by 6.8–41.6% over the three early-rice seasons. Combined inorganic/organic fertilizer application significantly increased CH_4 emissions, being average 95.7% and 82.9% greater for the FOM under ROM plots as compared with the control over the 3-year early-rice seasons, respectively (Tables 2 and 3).

There were similar temporal trends but varying in amplitudes of CH_4 fluxes among the plots with different fertilizer applications during the 2007–2009 late-rice growing seasons (Fig. 2). The CH_4 fluxes dramatically ascended after late-rice transplanting in July. The highest CH_4 fluxes were observed on days to 1 week after rice transplanting then gradually decreased to

Table 3 A two-way ANOVA for the effects of fertilizer (F) application and year (Y) on CH_4 and N_2O emissions and grain yields in rice paddies

			CH_4 (kg ha^{-1})			N_2O (kg ha^{-1})			Yield (t ha^{-1})		
Season	Factors	df	*SS*	*F*	*P*	*SS*	*F*	*P*	*SS*	*F*	*P*
Early-rice	F	5	252 182	9.99	<0.001	0.41	1.27	0.30	152.4	69.32	<0.001
	Y	2	106 874	10.58	<0.001	0.19	1.43	0.25	30.7	34.94	<0.001
	F × Y	10	26 527	0.53	0.86	0.57	0.88	0.56	6.7	1.53	0.17
	Model	17	385 584	4.49	<0.001	1.17	1.06	0.43	189.6	25.40	<0.001
	Error	36	181 828			2.34			15.8		
Late-rice	F	5	1 209 088	9.69	<0.001	1.63	7.32	<0.001	32.2	22.04	<0.001
	Y	2	47 486	0.95	0.40	0.13	1.41	0.26	16.9	28.94	<0.001
	F × Y	10	92 898	0.37	0.95	0.64	1.43	0.21	2.6	0.88	0.56
	Model	17	1 349 474	3.18	<0.01	2.40	3.16	<0.01	51.6	10.40	<0.001
	Error	36	898 309			1.61			10.5		
Non-rice season	F	5	68	5.43	0.01	61.59	22.74	<0.001			
	Y	2	66	2.09	0.11	58.39	8.62	<0.001			
	F × Y	10	211	3.38	0.007	81.92	6.05	<0.001			
	Model	17	365	3.43	0.003	175.09	7.61	<0.001			
	Error	24	150			32.5					

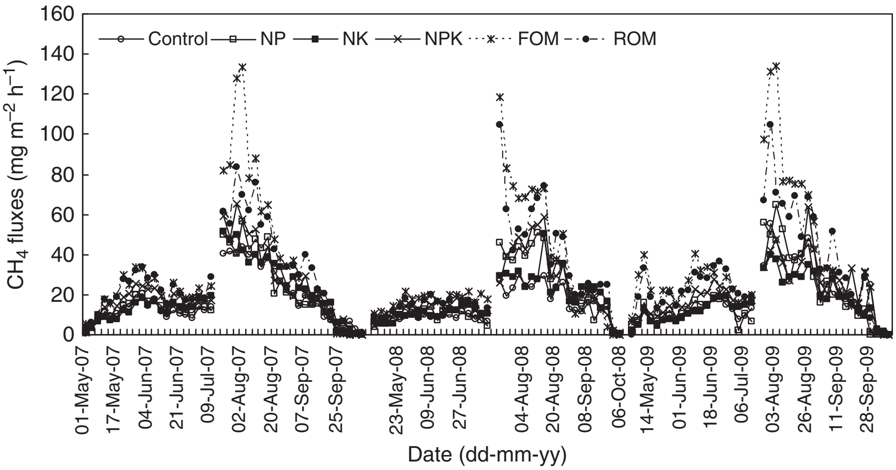

Fig. 2 Seasonal variation of methane (CH_4) fluxes from double rice-cropping systems from 2007 to 2009. Seasonal CH_4 fluxes in 2007 were redrawn from Yang *et al.* (2010).

background levels in the late-rice season. Particularly, a remarkable peak of CH_4 flux was observed approximately 1–2 weeks after late-rice transplanting for the plots with combined inorganic/organic fertilizer applications (Fig. 2). Substantial CH_4 emission was observed in the late-rice growing season, 113–157% greater than those in the early-rice season (Table 2, paired *t*-test, $P < 0.001$). Total CH_4 emission in the late rice-growing season depended greatly on fertilizer application, while it did not significantly vary with year and their interaction (Tables 2 and 3). Similar to the early-rice growing season, long-term combined inorganic/organic fertilizer (FOM and ROM) application significantly increased CH_4 emissions. Relative to the control, single inorganic fertilizers (NP, NK and NPK) tended to increase CH_4 emissions, although this effect was not statistically significant (Table 2).

In the nonrice winter seasons, all field treatment soils acted as small net sink or source of CH_4 to the atmosphere, which is largely due to drainage (Table 4). Annual CH_4 emissions, on average, ranged from 621 kg CH_4 ha^{-1} for the control to 1175 kg CH_4 ha^{-1} for the FOM plots over the three annual cycles. Compared with the control, the annual CH_4 emission was increased by 89.8% and 73.9% for the FOM and ROM plots, respectively. Long-term inorganic fertilizer (NP, NK and NPK) application in-

Table 4 Seasonal CH_4 (kg CH_4 ha^{-1}) and N_2O (kg N_2O–N ha^{-1}) emissions (Mean ± SD) during nonrice cropping winter seasons from rice paddies

	10/11/06–04/25/07*		10/18/07–05/01/08		10/14/08–04/27/09		2007–2009 winter seasons	
Treatment	CH_4	N_2O	CH_4	N_2O	CH_4	N_2O	CH_4	N_2O
Control	1.96	0.57	−0.99 ± 0.90	0.97 ± 0.29	0.67 ± 1.17	1.34 ± 1.25	−0.17 ± 1.43a	1.07 ± 0.80c
NP	3.01	3.62	0.07 ± 1.34	1.52 ± 0.45	−0.13 ± 0.81	0.67 ± 0.50	0.62 ± 1.47a	1.46 ± 1.11bc
NK	−2.59	2.83	−0.41 ± 1.00	2.55 ± 0.66	4.94 ± 1.02	2.57 ± 0.84	0.88 ± 3.34a	2.60 ± 0.63a
NPK	−4.55	2.16	−0.23 ± 0.41	1.37 ± 1.06	7.94 ± 4.92	1.50 ± 0.32	1.46 ± 5.91a	1.54 ± 0.69bc
FOM	−6.00	7.84	0.33 ± 0.50	2.23 ± 1.06	−2.88 ± 6.07	0.60 ± 0.30	−2.43 ± 4.26a	2.34 ± 2.65ab
ROM	1.63	4.94	0.24 ± 0.23	2.45 ± 0.76	2.06 ± 2.63	1.20 ± 0.57	0.58 ± 1.78a	2.27 ± 1.44ab

*mm/dd/yy, measurements were only taken from one plot in the 2006–2007 nonrice season.

Table 5 Mean annual CH_4 and N_2O emissions and soil organic carbon (SOC) sequestration rates and their estimated global warming potentials (GWPs) and greenhouse gas intensities (GHGIs) over the three annual cycle of 2006 winter season–2009 late rice-cropping season

Treatment	CH_4 (kg CH_4 ha^{-1} yr^{-1})	N_2O–N (kg N ha^{-1} yr^{-1})	SOCSR* (t C ha^{-1} yr^{-1})	Grain yield (kg ha^{-1} yr^{-1})	GWP†	GHGI‡
Control	622	1.15	0.96	6513	12 587	1.93
NP	685	2.46	1.33	9941	13 407	1.35
NK	650	2.97	1.01	6758	13 940	2.06
NPK	794	1.93	1.11	11 659	16 689	1.43
FOM	1174	4.11	1.42	12 841	26 066	2.03
ROM	1081	3.37	1.43	11 575	23 372	2.02

[b] *SOCSR* (t C ha^{-1} yr^{-1}) = $d_{SOC}/d_t \times \gamma \times (1-\delta_{2\,mm}/100) \times 20/10$; d_{SOC}/d_t (g C kg^{-1} yr^{-1}) was derived from the simulated logistic model for SOC increases with year as shown in Table 1, γ (g cm^{-3}) and $\delta_{2\,mm}$ represent the bulk density and the gravel content (>2 mm, 3–9%) of topsoil, soil tillage depth is 20 cm for rice paddies and the term 10 is the area conversion coefficient.

*Mean annual CH_4 or N_2O = early rice-cropping season + late rice-cropping season + winter season.

†GWP (kg CO_2-equivalent ha^{-1} yr^{-1}) = 25 × CH_4 + 298 × N_2O−44/12 × SOCSR.

‡GHGI (kg CO_2-equivalent kg^{-1} grain yield yr^{-1}) = GWP/grain yield of rice.

creased annual CH_4 emissions by 4.4–27.8% as compared with the control (Table 5).

N_2O emission

During the double-rice growing seasons, the paddy soils were typically small sink or source of N_2O for all field plots, except several pronounced fluxes due to fertilizer application and drainage (Fig. 3). Two-way ANOVA analyses indicated that seasonal amounts of N_2O emission during the rice-growing periods were not significantly affected by fertilizer, year and their interaction, except that N_2O emissions depended significantly on fertilizer application in the late-rice growing season (Tables 2 and 3). Over the 3-year early-rice seasons under continuous waterlogging, seasonal N_2O emissions from paddy fields were negligible, with an average of 2.18–11.17 μg N_2O–N m^{-2} h^{-1}, amounting to 0.04–0.20 kg N_2O–N ha^{-1} for various field experimental plots.

In contrast with continuous flooding in the early-rice season, several drainage episodes used in the late rice-growing season triggered much N_2O emission. Relative to the early-rice growing season, N_2O emissions were 13–98% greater in the late-rice season, except for no difference between the two rice-cropping seasons for the NK plots (Table 2). Over the three late-rice growing seasons, seasonal N_2O emissions from paddy fields averaged 2.60–18.11 μg N_2O–N m^{-2} h^{-1} for various field experimental plots, amounting to 0.06–0.41 kg N_2O–N ha^{-1} for each late rice-growing season. The highest N_2O emissions were observed from the FOM plots, significantly greater than those from the control and NP and NPK plots (Table 2).

Substantial N_2O emission was observed in the nonrice growing period, although no fertilizer was applied in the winter season (Table 4). Total N_2O emissions in the nonrice season varied significantly with fertilizer, year and their interaction (Table 3). Seasonal N_2O emissions were greater in the 2006–2007 nonrice winter

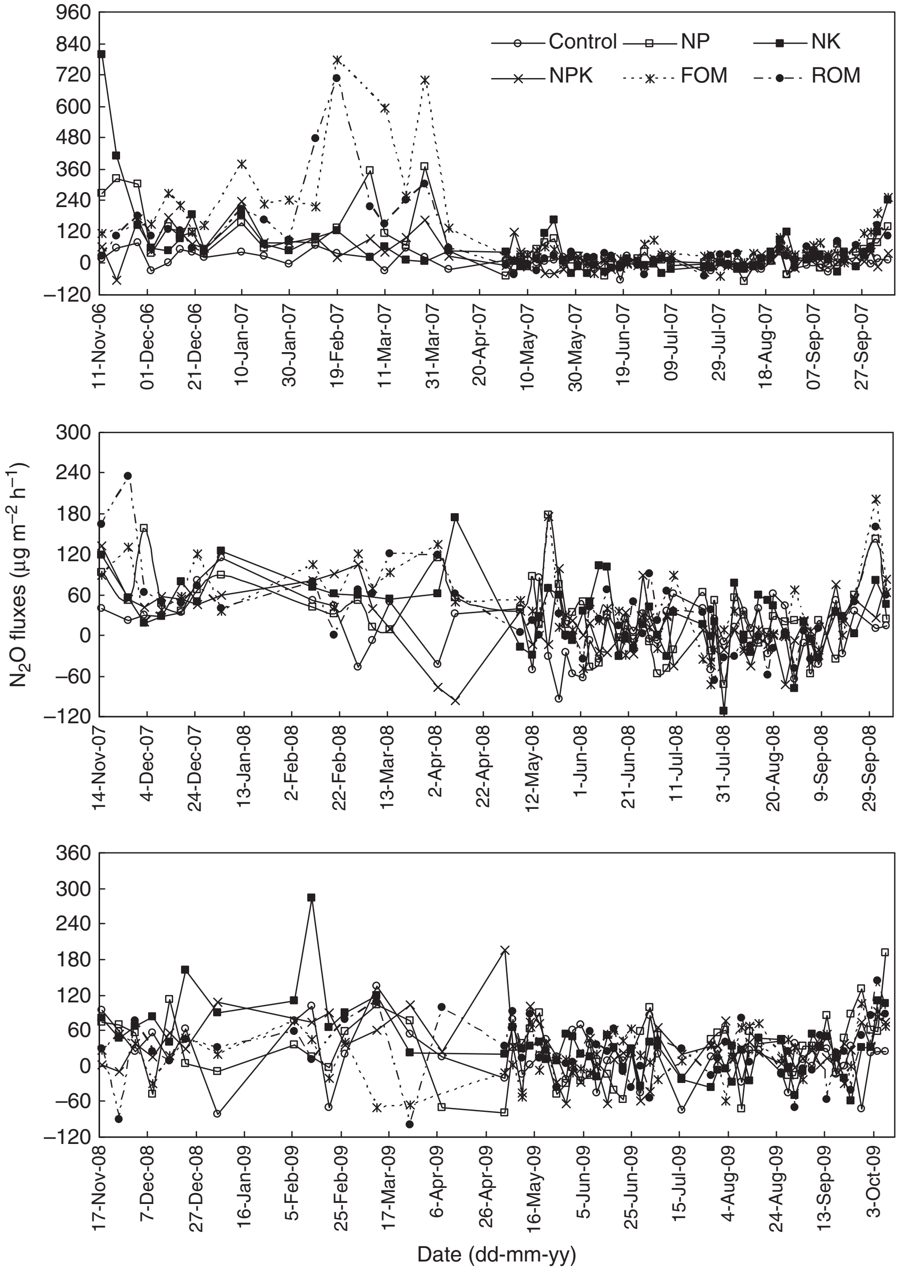

Fig. 3 Seasonal variation of nitrous oxide (N_2O) fluxes from double rice-cropping systems in three annual cycles over the period 2006–2009.

season than the other two winter seasons. Relative to the control, long-term fertilizer application in the rice-growing season significantly increased N_2O emissions in the nonrice winter season (Tables 3 and 4). Over the three nonrice growing seasons, seasonal total N_2O emissions averaged 1.07–2.60 kg N_2O–N ha^{-1}, with the lowest average flux of 22.27 μg N_2O–N m^{-2} h^{-1} for the control and the highest flux of 56.95 μg N_2O–N m^{-2} h^{-1} for the NK plots (Table 4).

Over the three annual rotation cropping cycles, annual total N_2O emissions ranged from 1.15 kg N_2O–N ha^{-1} for the control to 4.11 kg N_2O–N ha^{-1} for the FOM plots (Table 5). Compared with the control, annual N_2O emission was increased by 257% and 193% for the FOM and ROM plots, respectively. Long-term mineral fertilizer (NP, NK and NPK) application increased annual N_2O emissions by 68–158% relative to the control. Over the whole annual cycle, the emission factor of N_2O was, on average, estimated to be 0.43%, 0.72% and 0.99% due to the inorganic fertilizer NPK, NP and NK application, respectively. The emission factor of N_2O was estimated to be 0.09–0.17% and 0.09–0.24% for the inorganic fertilizer treatments during the early- and late-rice growing seasons, respectively.

Rice production

Grain yields of both early and late rice significantly varied with fertilizer and year, but independent of their interaction (Tables 2 and 3). Rice grain yields were greater in the 2007–2008 than in the 2009 season. Long-term fertilizer application significantly increased grain yields for the two rice-cropping systems, except for no significant difference between the NK and control plots. Compared with the control, organic/inorganic combination fertilizer application increased rice yields of early and late rice by 118–155% and 47–52% for the ROM and FOM plots over the three seasons, respectively. Rice yields of early and late rice were, on average, increased by 82–120% and 30–47% due to NP and NPK fertilizer application over the three growing seasons, respectively.

Net annual GWP and GHGI

Fertilizer application increased annual CH_4 and N_2O emissions, although it benefit atmospheric CO_2 sequestrated into soil, which led to an increase in the net GWPs over the 100-year time due to fertilizer application (Table 5). Compared with the control, inorganic fertilizer application increased the net annual GWPs by 7%, 11% and 32% for the NK, NP and NPK, respectively. The combined inorganic/organic fertilizer application greatly increased annual CH_4 and N_2O emissions, and thus the net annual GWPs were 86–107% greater for ROM and FOM plots relative to the control.

No significant difference in GHGIs was found between the NK and control, while the GHGIs were lower for the NP and NPK plots relative to the control since rice grain yields were significantly increased by NP and NPK fertilizer application (Table 5). Relative to the control, the inorganic fertilizer NP and NPK application decreased annual GHGIs by 26–30% over the three annual experimental cropping cycles. Compared with the control, in contrast, annual GHGIs were increased by 5% due to the organic/inorganic combination fertilizer application over the three annual experimental cropping cycles.

Discussion

Carbon sequestration in double rice-cropping systems

Net CO_2 exchange between the atmosphere and terrestrial systems represents the balance between C inputs by autotrophic fixation and outputs by heterotrophic oxidation of organic material. However, measurements of ecosystem CO_2 fluxes alone are not necessarily indicative of atmospheric loadings since it is difficult to partition soil heterotrophic respiration from soil CO_2 fluxes. In the present study, therefore, net ecosystem CO_2 balance was determined from changes in topsoil SOCD rather than ecosystem CO_2 fluxes.

The topsoil SOCD (36.4–48.2 $\mathrm{t\,C\,ha^{-1}}$) in the double rice-cropping systems in 2009 was slightly greater than previous estimates of regional or national mean SOCD of paddy soils based on earlier survey data summary (Pan *et al*., 2003; Huang & Sun, 2006; Xie *et al*., 2007; Liao *et al*., 2009). In contrast, Liu *et al*. (2006) used the increased accuracy of data sources (1 : 1000 000 soil database of China) and a larger profile data set (1490 paddy soil profiles) to estimate the SOCD of paddy soils in China. They estimated the mean SOCD to be 37.6 $\mathrm{t\,C\,ha^{-1}}$ for Chinese paddy topsoils and 39.6–52.7 $\mathrm{t\,C\,ha^{-1}}$ in our experimental region, which is highly comparable to the estimates of this study.

Consistent with previous reports, the double rice-cropping systems have experienced increases in topsoil SOC density over the last decade (Fig. 1). Annual increase rate of SOC averaged 0.49–0.80 $\mathrm{g\,C\,kg^{-1}\,yr^{-1}}$ in this study, which falls within the range of estimates based on long-term fertilizer experiment measurements (Wu & Cai, 2007). The SOC increase rate was as high as 1.03 $\mathrm{g\,C\,kg^{-1}\,yr^{-1}}$ in paddy soils under long term combined inorganic/organic fertilizer application in Yunnan over the period 1987–1998 (Wang, 2000). The SOC sequestration rate averaged 1.21 $\mathrm{t\,C\,ha^{-1}\,yr^{-1}}$ over the period 2007–2009 in the double rice-cropping paddy soils, generally greater than previous estimates in some single-rice paddy soils or upland soils (Li *et al*., 2006; Xie *et al*., 2007; Liao *et al*., 2009; Lu *et al*., 2009; Sun *et al*., 2009), although it falls within the SOC sequestration rate range of 0.13–2.20 $\mathrm{t\,C\,ha^{-1}\,yr^{-1}}$ estimated by Pan *et al*. (2003).

Several reasons may be given for greater SOCD and SOC sequestration rate in this study. First, grain yield averaged 6.5–12.8 $\mathrm{t\,ha^{-1}\,yr^{-1}}$ in this study, generally higher than that of annual rice-upland rotation systems or upland cropping systems (Huang *et al*., 2007). This suggests that greater SOCD is a consequence of increased crop residue retained into soils due to higher crop net primary production. Second, it could be attributed to surface waterlogging primarily dominated in double rice-cropping paddy soils, where the decomposition rate of SOC was slower than upland or single rice paddy-upland rotation systems. Furthermore, silt and clay contents in paddy soils are generally higher than those in upland soils, which also lead to larger SOC accumulation (Wang *et al*., 2005; Liu *et al*., 2006).

CH_4 emissions from double rice-cropping systems

Although CH_4 emissions from rice paddies have been well documented in the past decades, few measurements of CH_4 flux were taken in Chinese double rice-cropping systems (Wassmann *et al*., 1993; Lu *et al*., 2000; Qin *et al*., 2006; Xiong *et al*., 2007; Yang *et al*., 2010). In this study, annual CH_4 emissions, on average, varied between 622 and 1174 kg CH_4 ha^1 yr^{-1} over the 3 years (Table 5), within the upper ranges identified by Huang *et al*. (2004a) and Cai *et al*. (2000). Annual CH_4 emissions in this study were comparable to those previous results in rice paddies under continuous waterlogging, but much higher than those from other studies in the single rice-cropping systems with midseason drainage (e.g. Cai *et al*., 1997; Yan *et al*., 2005; Zou *et al*., 2005; Khalil *et al*., 2008). In the present study, continuous waterlogging was primarily dominated over the early-rice season. Although drainage was adopted in the late rice-cropping season, it was used in the late-stage rather than in the mid-season as usual. Midseason drainage of irrigated rice paddies often gives rise to a drop in seasonal CH_4 flux, and thereafter CH_4 flux hardly rebounds to the original level after reflooding (Cai, 1997; Mishra *et al*., 1997; Zou *et al*., 2005). In addition, cumulative CH_4 emissions during the late-rice seasons contributed most (63–83%) to the annual total of CH_4 emissions. The crop straw from the early rice were retained in the fields, providing a large addition of organic material under hot weather conditions favorable to quick decomposition during the late-rice cropping period (Huang *et al*., 2004a; Yan *et al*., 2005; Ma *et al*., 2009).

Effects of long-term fertilizer application on CH_4 emissions

It is generally believed that organic material incorporation increases CH_4 emissions from rice paddies (Adhya *et al*., 2000; Zou *et al*., 2005; Naser *et al*., 2007; Ma *et al*., 2009), which is supported by the evidence that CH_4 emissions were significantly higher from the FOM and ROM plots in this study. In contrast, previous reports on the influence of synthetic fertilizer on CH_4 emission from rice fields are inconsistent. Some studies showed that inorganic fertilizer application decreased CH_4 emissions in rice paddies (Wang *et al*., 1992; Cai *et al*., 1997; Krüger & Frenzel, 2003; Zou *et al*., 2009b), while increased CH_4 emission or no change in emission with inorganic fertilizer application was found in some other studies (Lindau *et al*., 1991; Wang *et al*., 1993; Cai *et al*., 2007). Nevertheless, the effects of inorganic fertilizer application on CH_4 emission are rarely examined in rice paddies under long-term fertilizer experiment, and thus these previous results did not reflect the effects of long-term inorganic fertilizer application on CH_4 emission. In the present study, the results of this study revealed that long-term inorganic fertilizer application significantly increased (early-rice season) or tended to increase CH_4 emissions (late-rice season) from rice paddies, which is probably associated with the increases in SOC content and crop biomass due to inorganic fertilizer application (Fig. 4). Significant linear relationships were found between seasonal CH_4 emission and the SOC content for each rice season during 2007–2009 (Fig. 4a), suggesting that long-term fertilizer application benefited SOC sequestration and thus stimulated CH_4 emissions from flooded rice paddies. Also, seasonal CH_4 emission depended greatly on crop growth, which was enhanced by inorganic fertilizer (Table 3, Fig. 4b).

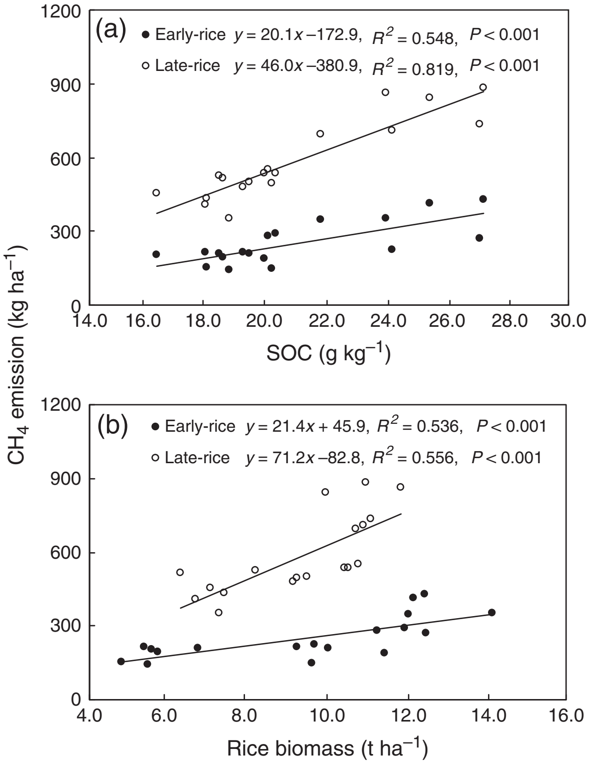

Fig. 4 Correlation between seasonal methane (CH_4) emission and soil organic carbon (SOC) content (a) and rice aboveground biomass (b) during the early- and late-rice seasons in 2007–2009. The slope of simulated linear relations differed between the two rice seasons.

Differences in seasonal CH_4 emission

Relative to the early-rice season, CH_4 emissions were significantly greater in the late-rice growing season. Several reasons may be given to the greater CH_4 emissions in the late-rice season. First, temperature was higher in the late-rice season than in the early-rice season (Appendix S1), which benefited soil CH_4 production. Second, the late-rice paddies have experienced longer period of flooding, and thus soil pH was lower after transplanting of the late-rice compared with that during the early-rice season, which created anaerobic soil environment suitable for CH_4 production (data not shown, but see Yang *et al.*, 2010). Third, CH_4 production depended largely on SOC content in the both rice-cropping seasons but differed in the slope of simulated linear relations, suggesting much more SOC decomposed into CH_4 in the late-rice than in the early-rice seasons (Fig. 4a). In addition, CH_4 production is related to Fe(III) reduction process in paddy soils. The paddy soils might contain considerable amounts of Fe(III) before the early-rice season. Given that Fe(III) might already have been reduced in the early-rice season under waterlogged conditions, the crop residues would provide the carbon source for the CH_4 pulse observed in the late-rice season (The *et al.*, 2008). Finally, rice cultivars were different between the two rice-cropping seasons. Rice plant serves as a main pathway of CH_4 emission, and the dependence of CH_4 emission on crop growth in rice paddies has been well documented (Huang *et al.*, 2004a; Yan *et al.*, 2005). Similar to the SOC, the dependence of CH_4 emission on rice biomass differed in the slope of linear relations, suggesting that the late-rice cultivars relative to the early-rice cultivars played more important roles in CH_4 emission (Fig. 4b).

N_2O emissions from double rice-cropping systems

Consistent with previous studies, N_2O emissions were negligible in flooded rice-cropping season (Akiyama *et al.*, 2005; Zou *et al.*, 2005; Liu *et al.*, 2010). Noted that much N_2O emission occurred in the nonrice period although no fertilizer was applied in the winter season, which might be due to several reasons. First, shifts from the anaerobic conditions prevailing in rice-cropping season to aerobic conditions after draining the fields benefit emissions of N_2O produced in soils, which resulted in a peak flux of N_2O during the fallow period after rice harvesting (Fig. 3). Second, flooding during the rice-growing season would create soil moisture more beneficial to N_2O production in the following nonrice season (Liu *et al.*, 2010). Finally, flooding lowered soil organic nitrogen mineralization during the rice season, but more mineral nitrogen in soil was available to N_2O production in the following nonrice season (Bouwman *et al.*, 2002; Yan *et al.*, 2003; Zheng *et al.*, 2004; Akiyama *et al.*, 2005; Liu *et al.*, 2010).

Emission factor and background emission of N_2O

The emission factor of N_2O was estimated to be 0.09–0.17% and 0.09–0.24% for the inorganic fertilizer treatments during the early- and late-rice growing seasons, respectively, comparable to previous estimates in the flooded rice paddies (Akiyama *et al.*, 2005; Zou *et al.*, 2005, 2007). Over the whole annual cycle, the emission factor of N_2O was, on average, estimated to be 0.43%, 0.72% and 0.99% due to the inorganic fertilizer NPK, NP and NK application, respectively. Our previous studies in an annual rice–winter wheat rotation system showed that the emission factor of N_2O averaged 0.89% over the whole annual cycle (Liu *et al.*, 2010). However, the values of annual emission factors in this study are generally lower than those previous estimates in Chinese upland croplands (Yan *et al.*, 2003; Zheng *et al.*, 2004).

Annual N_2O emissions from the control without fertilizer application represent the background emission of N_2O, with an average of $1.15\,\mathrm{kg\,N_2O–N\,ha^{-1}}$, comparable to previous reports (1.23 ± 0.23) in a rice-based cropping system experienced continuous waterlogging in the rice-growing season (Liu *et al.*, 2010). By summary, the available data on field measurements, Gu *et al.* (2009) reported annual background N_2O emissions with an range of $1.14–2.53\,\mathrm{kg\,N_2O–N\,ha^{-1}}$ in Chinese rice-based cropping rotations. Nevertheless, background N_2O emissions may be dependent on SOC and total nitrogen contents, bulk density and clay fraction (Gu *et al.*, 2007).

Net GWP and GHGI of double rice-cropping production

The net GHGIs of this study is generally lower than the DNDC model estimates ($3.22\,\mathrm{kg\,CO_2}$-equivalents $\mathrm{kg^{-1}}$-grain yield $\mathrm{yr^{-1}}$) in continuous waterlogging rice paddies, but higher than our previous estimates (0.24–0.74 $\mathrm{kg\,CO_2}$-equivalents $\mathrm{kg^{-1}}$ grain yield $\mathrm{yr^{-1}}$) of rice paddies with midseason drainage and organic manure incorporation (Li *et al.*, 2006; Qin *et al.*, 2010), which is largely due to the rice season dominated by waterlogging. Therefore, midseason drainage instead of continuous waterlogging should be adopted to greatly reduce CH_4 emissions, and thus alleviates the climatic impacts of double rice-cropping production. Although midseason drainage results in a trade-off between CH_4 and N_2O emissions, flooding-midseason drainage-reflooding has been suggested to be an effective water regime for mitigating the net GWPs of CH_4 and N_2O emissions from rice paddies (Li *et al.*, 2005, 2006; Zou *et al.*, 2005). Also, some studies showed that poor drainage during winter drained season incurred a high CH_4 emission in flooded rice season (Xu *et al.*, 2002, 2003), suggesting water management over the whole annual cycle plays an important role in terms of reducing annual CH_4 emission from rice paddies.

Compared with the control, inorganic fertilizer application slightly increased the net annual GWPs, whereas the inorganic fertilizer NP and NPK application decreased annual GHGIs by 23–29%. Based on the results of grain yield, GWPs and GHGIs, balanced fertilizer application, particularly P fertilizer supplement, would be used to simultaneously achieve grain yield and mitigating climatic impacts of double rice-cropping production. Relative to the control, in contrast, combined inorganic/organic fertilizer application greatly increased the net annual GWPs and GHGIs over the three annual experimental cropping cycles. In order to avoid much CH_4 emission triggered by organic material amendment, therefore, organic material such as crop residue is recommended to be incorporated into soil in the drained nonrice growing season rather than the flooded rice-cropping season (Ma *et al*., 2009; Yang *et al*., 2010). Indeed, crop residues with high C : N ratio incorporated into soil in the nonrice growing season would not only benefit soil SOC accumulation, but also reduce N_2O emissions (Huang *et al*., 2004b; Zou *et al*., 2004).

Fertilizer management strategy in double rice-cropping systems

In the present study, grain yield was comparable between the control and NK treatments, while it was significantly higher for the NP and NPK plots. This suggests that phosphorus fertilizer is crucial to achieving high yield of double rice production, which is supported by the lower soil available P content for the NK plots relative to the other fertilizer applied plots (Appendix S2). Therefore, P fertilizer supplement is essential to increasing soil nutrient availability and rice production since the soil is generally P deficit due to the acidic soil condition. Indeed, soil available P content is generally lower in this long-term experimental double rice-cropping system than in other long-term fertilizer experimental systems in China (Zhao *et al*., 2010). In contrast, grain yields of rice were greater for the NPK and NP plots relative to the control, suggesting that K fertilizer might be not as important as P fertilizer to the double-rice production. Compared with other long-term fertilizer experiments in China, soil available potassium content was comparable or higher for all the fertilizer treatments in this study (Zhao *et al*., 2010). This is probably due to much K derived from crop residue retained in the fields.

Conclusions

Taking effective agriculture management strategy for mitigating climatic impacts requires a complete perspective on the agriculture impacts on radiative forcing. This study provided an insight into a complete GHG accounting of GWP and GHGI as affected by long-term fertilizer application in typical double rice-cropping systems. Long-term fertilizer application significantly increased grain yields and topsoil SOC density, except for no difference between the control and NK plots. Inorganic fertilizer application tended to increase CH_4 emissions during the rice-growing season and significantly increased N_2O emissions from drained soils during the nonrice season. Long-term combined inorganic/organic fertilizer application remarkably increased the net annual GWPs, largely due to CH_4 emissions under continuous waterlogging. The GHGI was lowest for the NP and NPK plots, and highest for the FOM and ROM plots. In order to simultaneously achieve high grain yield and low GHGI, we proposed some agricultural management strategies in the double rice-cropping systems, including balanced fertilizer management (particularly P fertilizer supplement), midseason drainage instead of continuous waterlogging and crop residue incorporated into soil in the non-rice drained season rather than in the flooded rice season.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Appendix S1. Monthly mean air temperature and precipitation during the experimental seasons over the period 2006–2009 at the long-term fertilizer experiment station.
Appendix S2. Soil physiochemical properties in the experimental fields under long-term different fertilizer treatments in Taoyuan, Hunan, China.
Appendix S3. Fertilizer application regime treatments in double rice-cropping fields at long-term experimental station from 2007–2009.