

## CROPS AND SOILS RESEARCH PAPER

# Methane and nitrous oxide emissions as affected by long-term fertilizer management from double-cropping paddy fields in Southern China

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### SUMMARY

There is limited information about the influences of long-term fertilizer management on methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) emissions from double-cropping paddy fields in Southern China. Therefore, the objective of the present study was to characterize the changes of CH<sub>4</sub> and N<sub>2</sub>O related to different fertilizer treatments based on a long-term field experiment. The experiment was initiated in 1986 and consisted of five treatments: unfertilized (CK), mineral fertilizer alone (MF), rice residues plus mineral fertilizer (RF), low manure rate plus mineral fertilizer (M1 + F), and high manure rate plus mineral fertilizer (M2 + F). Investigations were conducted over 2 years, from 2013 to 2014, to examine the CH<sub>4</sub> and N<sub>2</sub>O emissions from paddy field of Southern China. The results indicated that M2 + F plots had the largest CH<sub>4</sub> emissions during the early rice and late cropped rice and that MF and RF had larger N<sub>2</sub>O emissions than CK in both early and late cropped rice. When compared with the control, total N<sub>2</sub>O emissions in both rice-growing seasons increased in both MF and RF in 2013 and 2014. The global warming potentials (GWP) from paddy fields were ranked as M2 + F > M1 + F > RF > MF > CK. Meanwhile, the results demonstrated that CH<sub>4</sub> and N<sub>2</sub>O emissions were closely associated with the soil redox potential and soil temperature. In summary, the incorporation of rice residues in addition to the use of mineral fertilizer (RF treatment) may be an effective fertilizer management practice for mitigating total GWP per grain yield and maintaining rice grain yield in southern China.

### INTRODUCTION

With the current rise in global temperature, numerous studies have focused on greenhouse gas (GHG) emissions (Levy *et al.* 2007; Sagar *et al.* 2007; Hernandez-Ramirez *et al.* 2009). In addition to carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) play important roles in global warming. The global warming potentials (GWP) of CH<sub>4</sub> and N<sub>2</sub>O are 25 and 298 times that of CO<sub>2</sub> in a time horizon of 100 years, respectively (Bhatia *et al.* 2005) and their concentrations in the atmosphere are estimated to be increasing at the rates of 1 and 0.2–0.3% per year (Verge *et al.* 2007), respectively. In addition to industrial emissions, farmland and agricultural production are major sources of GHG emission (Wassmann *et al.* 2004; Lokupitiya & Paustian 2006; Verma

*et al.* 2006; Liu *et al.* 2008; Tan *et al.* 2009). Numerous findings have indicated that rice (*Oryza sativa* L.) paddy fields are a significant source of CH<sub>4</sub> and N<sub>2</sub>O emissions (Tan *et al.* 2009; Kallenbach *et al.* 2010). Thus, the characteristics of CH<sub>4</sub> and N<sub>2</sub>O emissions from paddy fields and the reduction of these emissions has received attention from scientists.

A considerable number of studies have shown that some farm operations can influence CH<sub>4</sub> and N<sub>2</sub>O emissions. For example, cropping system, crop type, water and nitrogen (N) management, organic matter application and tillage can help to regulate CH<sub>4</sub> and N<sub>2</sub>O emissions (Yagi & Minami 1990; Akiyama & Tsuruta 2002; Al-Kaisi & Yin 2005). Cropping system can affect the quality and quantity of crop residues returned to the soil, and eventually influence CH<sub>4</sub> and N<sub>2</sub>O emissions (Mosier *et al.* 2006; Sainju *et al.* 2008). Tillage and crop residue (straw) retention have a tremendous influence on CH<sub>4</sub> and N<sub>2</sub>O

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emission through the alteration of soil properties (e.g. porosity, temperature, moisture, etc.) (Al-Kaisi & Yin 2005; Yao *et al.* 2009). In paddy soils, CH<sub>4</sub> is produced by archaea bacteria during the anaerobic degradation of organic matter and oxidized by methanotrophic bacteria (Groot *et al.* 2003). Incorporation of organic material into the soil can enhance numbers of archaea bacteria and their activity (Yue *et al.* 2005), and provide large quantities of active organic substrate for CH<sub>4</sub> production (Sethunathan *et al.* 2000). Soil amendment with organic material, such as crop residue (Ma *et al.* 2008) and manure incorporation (Wang *et al.* 2013), has been estimated to promote CH<sub>4</sub> emission in paddy fields. Biogenic N<sub>2</sub>O production originates from nitrification and denitrification (Chu *et al.* 2007), which are processes involving soil microorganisms. Some studies have indicated that N fertilization usually stimulates N<sub>2</sub>O emissions (Mosier *et al.* 2006; Dusenbury *et al.* 2008; Robertson & Vitousek 2009).

Nitrous oxide and CH<sub>4</sub> emissions are indirectly affected by soil temperature and redox potential (Parkin & Kaspar 2003; Dusenbury *et al.* 2008; Liebig *et al.* 2010). The emission of N<sub>2</sub>O and CH<sub>4</sub> has been shown to depend on the atmospheric N-input rate, soil temperature, water content, and the chemical and physical characteristics of the soil (Vor *et al.* 2003). Similarly, cropping sequence and crop species can influence soil temperature by affecting shade intensity and evapotranspiration (Curtin *et al.* 2000; Amos *et al.* 2005). Nitrogen fertilization can reduce soil temperature compared with no N fertilization by increasing shade intensity through increased biomass production (Sainju *et al.* 2008).

The middle and lower Yangtze River Plain is one of the most vital rice production bases in China. Since the 1980s, traditional fertilizer management practices have been altered considerably (Du *et al.* 2009; Tang *et al.* 2014). With the continuous increase of mineral fertilizer application rates, manure inputs have been declining dramatically. Meanwhile, returning crop residue to the field is being accepted gradually. There is a growing concern that the increase of mineral fertilizer application rates may be unsustainable due to their promoting soil CH<sub>4</sub>, N<sub>2</sub>O emission and decreasing crop yield. The effect of mineral fertilizer alone on CH<sub>4</sub> and N<sub>2</sub>O emissions under a barley–double-cropping rice system is still undocumented. In addition, applications of crop residue and manure are viable options for maintaining crop yield, but it is important to monitor CH<sub>4</sub> and N<sub>2</sub>O emissions and related microbial activities.

Therefore, the objectives of the current research were to quantify CH<sub>4</sub> and N<sub>2</sub>O emissions from a paddy field and rice grain yield with long-term application of crop residue, manure and mineral fertilizer in barley–double-cropping rice systems in Southern China.

## MATERIALS AND METHODS

### Sites and cropping system

The experiment was established in 1986. It was conducted in Ning Xiang County (28°07'N, 112°18'E; 36 m asl) of Hunan Province, China. Under a continental monsoon climate, the annual mean precipitation is 1553 mm and potential evapotranspiration of 1354 mm. The monthly mean temperature is 17.2 °C. Soil texture in the plough layer (0–20 cm) was silt clay loam with 13.71% sand and 57.73% silt. At the beginning of the study, the surface soil characteristics (0–20 cm) were as follows: soil organic carbon (SOC) 29.4 g/kg, total N 2.0 g/kg, available N 144.1 mg/kg, total phosphorous (P) 0.59 g/kg, available P 12.87 mg/kg, total potassium (K) 20.6 g/kg, and available K 33.0 mg/kg. There were three crops in a year, barley (*Hordeum vulgare* L.), early rice and late rice. Barley was sown in the middle of November and harvested in early May of the following year. Early rice was then transplanted and harvested in the middle of July. The growing season of late transplanted rice lasted from late July to the end of October.

### Experimental design

The experiment had five treatments: control (without fertilizer input, CK), mineral fertilizer only (MF), rice residue plus mineral fertilizer (RF), low manure rate plus mineral fertilizer (M1 + F), and high manure rate plus mineral fertilizer (M2 + F). The design ensured all fertilized treatments received the same N rate (the amount of N in mineral fertilizer plus that from rice residue or manure) of 142.5 kg/ha during the early rice growing season and 157.5 kg/ha during the late rice growing season. All the fertilized treatments received the same amount of phosphorus pentoxide (P<sub>2</sub>O<sub>5</sub>), including the amount of P<sub>2</sub>O<sub>5</sub> in mineral fertilizer plus that from rice residue or manure: 54.0 kg/ha during the early rice growing season and 43.2 kg/ha during the late rice growing season. Likewise, all the fertilized treatments received the same amount of potassium oxide (K<sub>2</sub>O), constituting the amount of K<sub>2</sub>O in mineral fertilizer plus that from rice residue or manure:

63.0 kg/ha during the early rice growing season and 81.0 kg/ha during the late rice growing season. The mineral fertilizers included urea, ordinary superphosphate and potassium chloride. Details about the fertilizer management are listed in Table 1. Before transplanting rice seedlings, air-dried rice residue was manually spread onto the soil surface and incorporated into the soil at a cultivation depth of c. 20 cm. For early and late cropped rice, 40 and 30%, respectively, of mineral N fertilizer was applied at seeding and the remaining N fertilizer was applied by top dressing (7–10 days after transplanting) during crop growth. All the P and K fertilizers were applied at seeding. There were three replications and each plot size was 66.7 m<sup>2</sup> (10 × 6.67 m<sup>2</sup>). Data for the individual cropping periods investigated in the present study are referred to as 2013 and 2014, respectively.

Collection and measurement of methane and nitrous oxide

Methane and N<sub>2</sub>O emitted from paddy fields were collected using the static chamber technique at 09:00–11:00 h during the early rice and late rice growing seasons. The chamber (80 × 80 × 120 cm<sup>3</sup>) was made of 5-mm polyvinyl chloride (PVC) board and a PVC base with a collar. The base had a groove in the collar, into which the sides of the chamber were settled, and inserted into the soil c. 5 cm deep with nine rice plants growing inside the base. The groove was 1 cm below the level of the flooded water, and the sides of the chamber were settled into the groove of the collar with water to prevent leakage and gaseous exchange. The chamber contained a small fan for circulating the air, a thermometer sensor and a triple-vent hole. From the second day after transplanting of early or late rice, gases were sampled weekly. Before sampling, the fan in the chamber was switched on to allow even air mixture before extracting the air with a 50-ml syringe at 0, 10, 20 and 30 min after closing the box. The air samples were transferred into 0.5-litre sealed sample bags by rotating the trinal vent hole.

The quantities of CH<sub>4</sub> and N<sub>2</sub>O emission were measured with a gas chromatograph (Agilent 7890A, Agilent Technologies Co., Ltd., CA, USA) equipped with flame ionization detector (FID) and electron capture detector (ECD). Methane was separated using a 2-m stainless-steel column with an inner diameter of 2 mm, 13XMS column (60/80 mesh), with FID at 200 °C. Nitrous oxide was separated using a 1-m

Table 1. Nutrient supply\* from rice straw, chicken manure and mineral fertilizer under different fertilizer treatments

Treatment†	Early rice			Late rice			Total		
	N	P <sub>2</sub> O <sub>5</sub>	K <sub>2</sub> O	N	P <sub>2</sub> O <sub>5</sub>	K <sub>2</sub> O	N	P <sub>2</sub> O <sub>5</sub>	K <sub>2</sub> O
CK	0+0	0+0	0+0	0+0	0+0	0+0	0	0	0
MF	142.5+0	54.0+0	63.0+0	157.5+0	43.2+0	81.0+0	300.0	97.2	144.0
RF‡	124.4+18.1	50.4+3.6	38.3+24.7	133.0+24.5	37.8+5.4	48.2+32.8	300.0	97.2	144.0
M1+FS	96.0+46.5	33.0+21.0	33.6+29.4	110.2+47.3	21.8+21.4	51.1+29.9	300.0	97.2	144.0
M2+F#	49.6+92.9	12.0+42.0	4.2+58.8	63.0+94.5	0.5+42.7	21.2+59.8	300.0	97.2	144.0

N, nitrogen; P<sub>2</sub>O<sub>5</sub>, phosphorus pentoxide; K<sub>2</sub>O, potassium oxide; CK, without fertilizer; MF, mineral fertilizer alone; RF, crop residue plus mineral fertilizer; M1 + F, low manure rate plus mineral fertilizer; M2 + F, high manure rate plus mineral fertilizer.

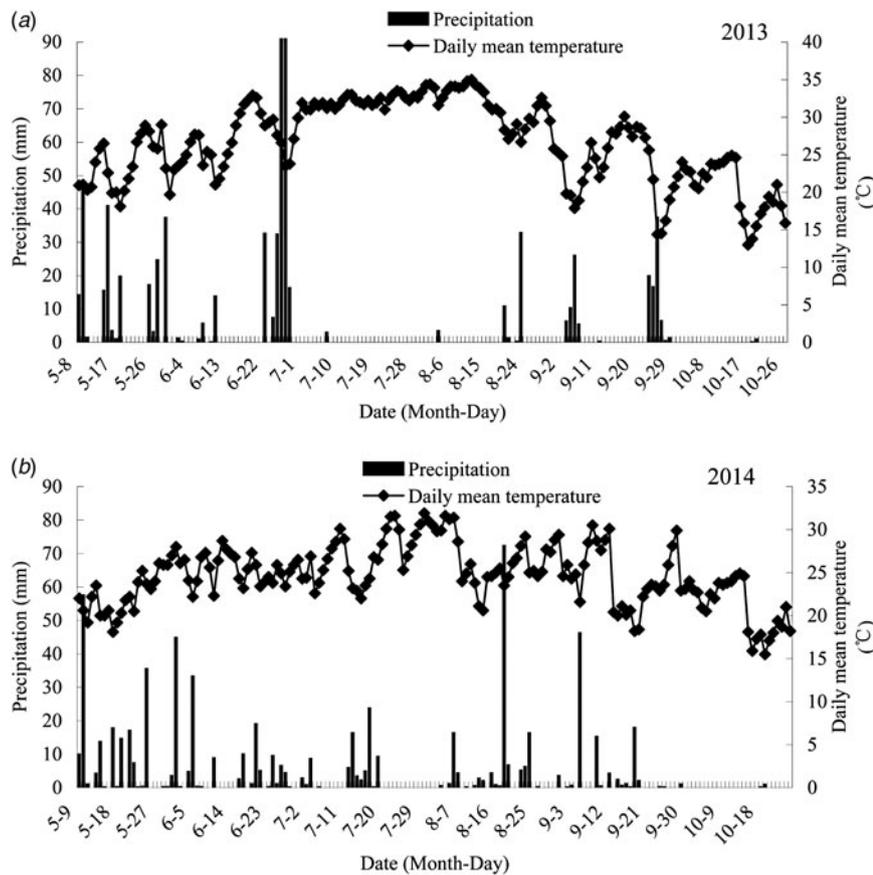
\* Input from mineral fertilizer + input from organic fertilizer. The numbers are in kg/ha.

† The N, P and K content of air-dry early rice straw was 6.5, 1.3 and 8.9 g/kg, N, P and K content of air-dry late rice straw was 6.8, 1.5 and 9.1 g/kg, respectively, and N, P and K content of decomposed chicken manure was 17.7, 8.0 and 11.2 g/kg, respectively.

# For the RF treatment, the rice straw return rate (air dry) was 2780 and 3600 kg for early and late rice.

§ For the M1 + F treatment, the manure application rate (decomposed) was 2625.0 and 2670.0 kg for early and late rice.

# For the M2 + F treatment, the manure application rate (decomposed) was 5250.0 and 5340.0 kg for early and late rice.



**Fig. 1.** Daily precipitation and mean temperature during the experimental period at the experimental site.

stainless-steel column with an inner diameter 2 mm, Porapak Q (80/100 mesh) and ECD at 330 °C.

Methane and N<sub>2</sub>O fluxes were calculated with the following equation (Liebig *et al.* 2010):

$$F = \rho h [273 / (273 + T)] dC / dt$$

where  $F$  was the CH<sub>4</sub> flux (mg/m<sup>2</sup>/h) or N<sub>2</sub>O flux (μg/m<sup>2</sup>/h);  $T$  was the air temperature (°C) inside the chamber;  $\rho$  was the CH<sub>4</sub> or N<sub>2</sub>O density at standard state (0.714 kg/m<sup>3</sup> for CH<sub>4</sub> and 1.964 kg/m<sup>3</sup> for N<sub>2</sub>O);  $h$  was the headspace height of the chamber (m); and  $dC/dt$  was the slope of the curve of gas concentration variation with time.

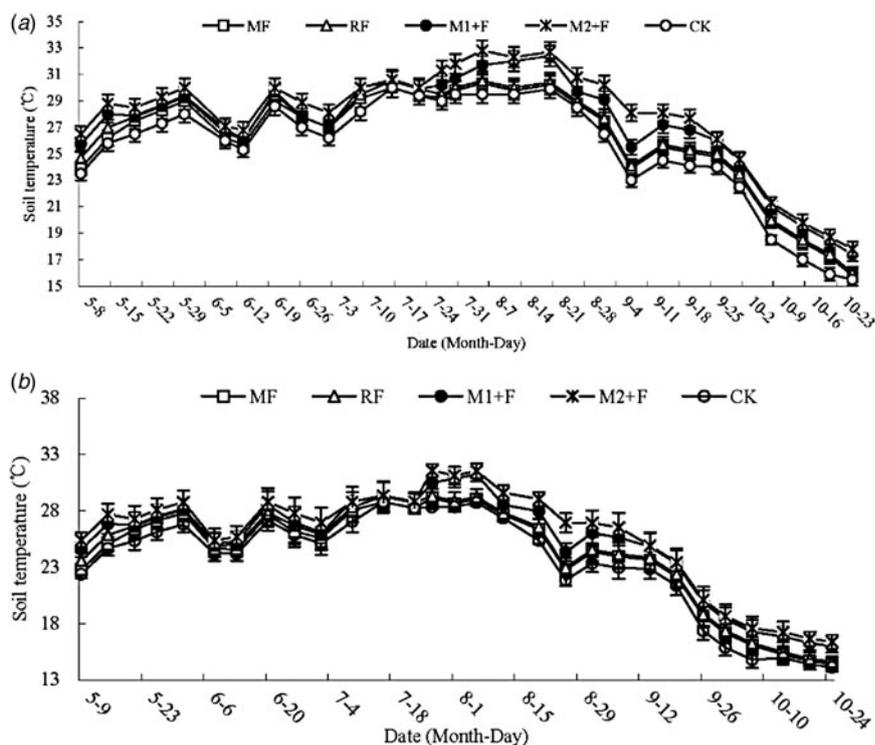
The total CH<sub>4</sub> and N<sub>2</sub>O emissions were sequentially computed from the emissions between every two adjacent intervals of measurements, based on a non-linear, least-squares method of analysis (Parashar *et al.* 1993; Singh *et al.* 1996).

Global warming potential was defined as the cumulative radiation forcing both direct and indirect effects integrated over a period of time from the emission of a unit mass of gas relative to some reference gas.

Carbon dioxide was chosen as the reference gas. The GWP conversion parameters of CH<sub>4</sub> and N<sub>2</sub>O (over 100 years) were adopted with 25 and 298 kg CO<sub>2</sub>-equivalent/ha (IPCC 2013).

#### Investigation of soil properties

Soil redox potential (Eh value) and soil temperature were periodically recorded along with gas collection throughout the early rice and late rice cultivation period. The Eh electrode was installed permanently at 10 cm soil depth and measured in each plot using an Eh meter (PRN-40, Fuji-wara Scientific, Japan). Soil temperature was recorded with a thermometer (PC-9125, AS ONE Co., Tokyo, Japan) installed at 10 cm soil depth. Air temperature and rainfall data were obtained from Ning Xiang Meteorological Observatory of Changsha Meteorological Station, China (2.5 km east of the experimental site). The daily precipitation and mean temperature during the experimental period are shown in Fig. 1. The temporal variations of the soil temperature and soil Eh at 10 cm



**Fig. 2.** Soil temperature at 10 cm depth in paddy field during early and late cropped rice in (a) 2013 and (b) 2014. MF, mineral fertilizer alone; RF, crop residue + mineral fertilizer; M1 + F, low manure rate + mineral fertilizer; M2 + F, high manure rate + mineral fertilizer; CK, without fertilizer. Bars show s.d.

deep during the experimental period are shown in Figs 2 and 3.

#### Other measurements

Grain yield was determined from a 1 m<sup>2</sup> sampling area at harvest and was expressed as rough (unhulled) rice at 14% moisture content. Above-ground straw yield was determined after drying plant material at 80 °C for 2 days.

#### Statistical analysis

All data were expressed as mean  $\pm$  s.e.. The data were analysed as a randomized complete block, using the PROC ANOVA procedure of SAS (SAS Institute 2003). Mean values were compared using the least significant difference test, significant at  $P < 0.05$ .

## RESULTS

### Methane emission

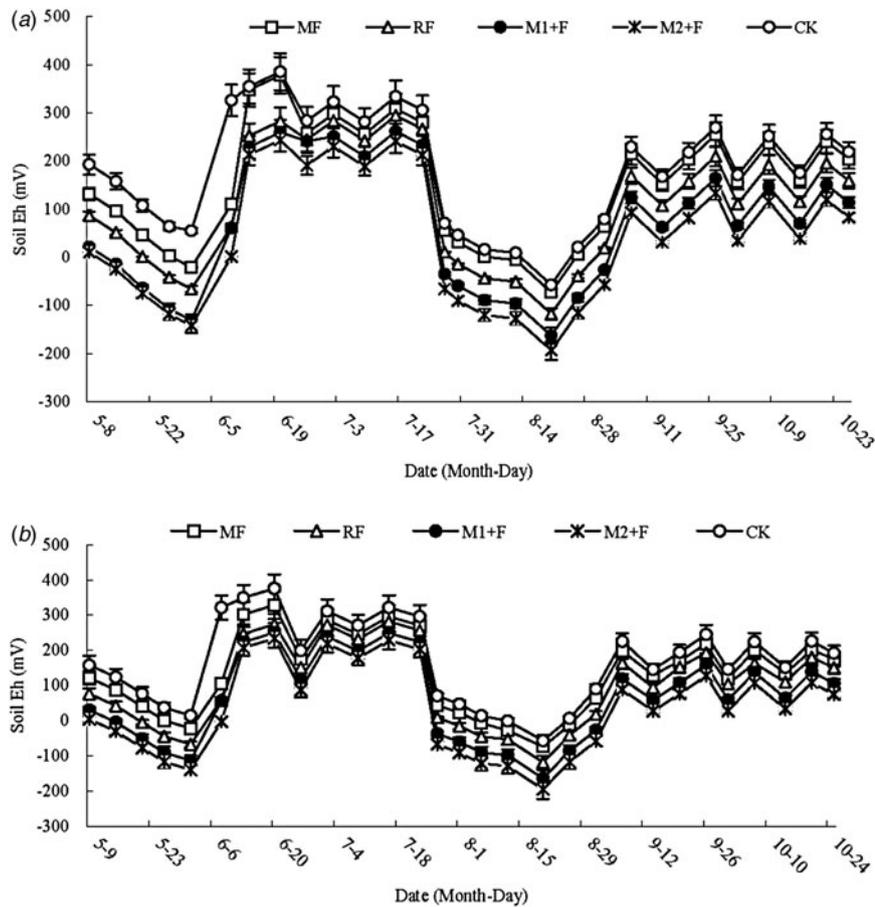
For the early rice crop the curve of CH<sub>4</sub> flux was low following transplanting but increased quickly until the peak

appeared at 24 and 23 days after transplanting in 2013 and 2014, respectively. In both the seasons, there then followed a dramatic decline to a low level (Fig. 4). In the early rice season, the CH<sub>4</sub> flux values were significantly different among treatments of the order of M2 + F > M1 + F > RF > MF > CK ( $P < 0.05$ ) (Fig. 4).

Methane emission in the late rice crop was focused mainly at the tillering stage (growth stage (GS) 21–29; Zadoks *et al.* 1974), and the peak value of CH<sub>4</sub> flux was observed at 25 and 24 days after transplanting in all treatments in both years. The emission rate then decreased dramatically to a low and stable level, especially from field drainage to harvest. The order of treatments in CH<sub>4</sub> emission was the same as for the early rice crop. (Fig. 4).

### Nitrous oxide emissions

The peak flux of N<sub>2</sub>O was emitted when the field was drained. Meanwhile, some N<sub>2</sub>O was also emitted during the alternating wetting–drying irrigation period. The first peak value of N<sub>2</sub>O flux appeared at 7 days after transplanting in all treatments in both 2013 and 2014, before decreasing. The order among treatments was MF > M2 + F > M1 + F > RF >



**Fig. 3.** Soil redox potential (Eh) at 10 cm depth in paddy field during early and late cropped rice in (a) 2013 and (b) 2014. MF, mineral fertilizer alone; RF, crop residue + mineral fertilizer; M1 + F, low manure rate + mineral fertilizer; M2 + F, high manure rate + mineral fertilizer; CK, without fertilizer. Bars show s.d.

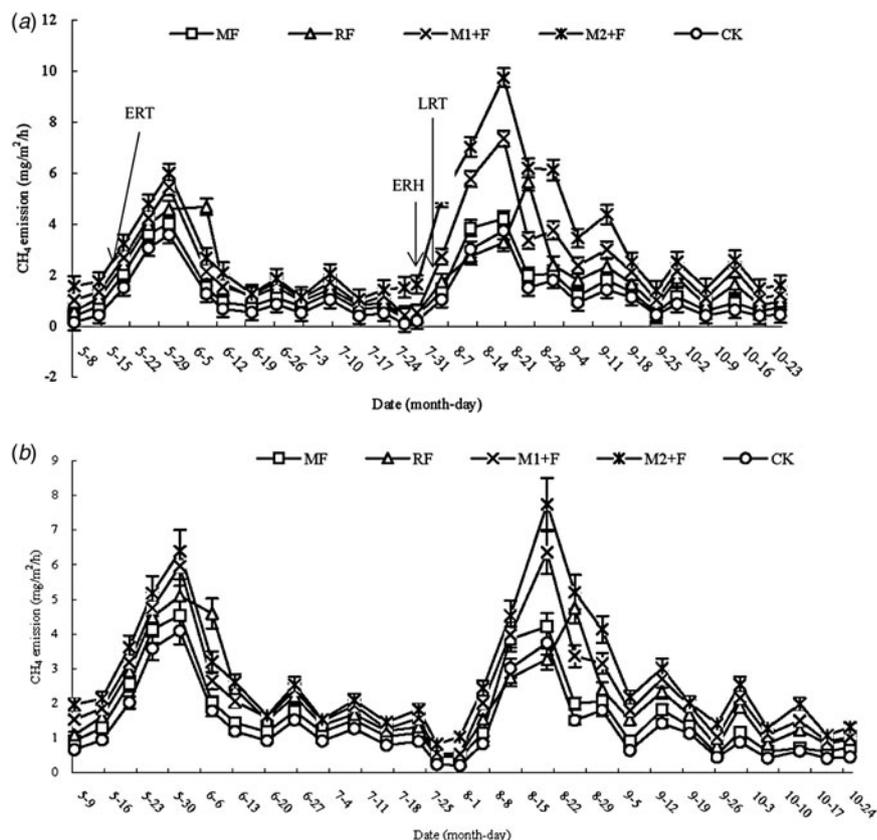
CK during the period from transplanting to field drainage, and  $RF > M2 + F > M1 + F > M + F > CK$  during the alternating wetting–drying irrigation period. The N<sub>2</sub>O flux in early paddy rice reached the highest peak at 37 and 35 days after transplanting in 2013 and 2014, respectively (Fig. 5).

In the late rice crop, N<sub>2</sub>O emissions increased from field drainage to full heading stage (GS 50–60), and was mainly focused at the booting stage (GS 41–47). The order of N<sub>2</sub>O emission fluxes among different treatments was  $MF > M2 + F > M1 + F > RF > CK$  during the period from transplanting to field drainage, and  $RF > M2 + F > M1 + F > MF > CK$  during the alternating wetting–drying irrigation period. In 2013, the average N<sub>2</sub>O fluxes in the late rice growing season were 20.13, 20.37, 15.44, 17.98 and 11.68  $\mu\text{g}/\text{m}^2/\text{h}$  in MF, RF, M1 + F, M2 + F and CK, respectively. In 2014, the average N<sub>2</sub>O fluxes in the late rice growing season were 21.05, 21.33, 18.46, 20.50 and 14.68  $\mu\text{g}/\text{m}^2/\text{h}$  in MF, RF, M1 + F, M2 + F and CK, respectively.

Total methane and nitrous oxide emission from paddy fields of early rice and late rice

The cumulative CH<sub>4</sub> emission of CK was significantly lower than MF, RF, M1 + F and M2 + F during the early rice crop ( $P < 0.05$ ), and the sequence of treatments was  $M2 + F > M1 + F > RF > MF > CK$  (Table 2). In 2013, the total CH<sub>4</sub> emissions from paddy fields covering the whole late rice growth cycle were 3.35, 4.27, 5.86, 8.43 and 2.65  $\text{g}/\text{m}^2$  in MF, RF, M1 + F, M2 + F and CK, respectively. In 2014, the total CH<sub>4</sub> emissions from paddy fields during the whole late rice growth cycle were 3.21, 3.96, 4.88, 6.10 and 2.55  $\text{g}/\text{m}^2$  in MF, RF, M1 + F, M2 + F and CK, respectively. The sequence of treatments with respect to total CH<sub>4</sub> emission was  $M2 + F > M1 + F > RF > MF > CK$  (Table 2).

Compared with CK, other treatments increased total N<sub>2</sub>O emissions in the early rice crop with emissions increasing by 0.008  $\text{g}/\text{m}^2$  (88.89%) in MF, 0.007  $\text{g}/\text{m}^2$  (77.78%) in RF, 0.005  $\text{g}/\text{m}^2$  (55.56%) in M1 + F and



**Fig. 4.** Methane ( $\text{CH}_4$ ) fluxes from the soils affected by long-term fertilizer management during early and late cropped rice in (a) 2013 and (b) 2014. MF, mineral fertilizer alone; RF, crop residue + mineral fertilizer; M1 + F, low manure rate + mineral fertilizer; M2 + F, high manure rate + mineral fertilizer; CK, without fertilizer; ERT, early rice transplanting; ERH, early rice harvesting; LRT, late rice transplanting. Methane emission rate is the mean of values measured within each treatment ( $n = 3$ ). Bars show S.D.

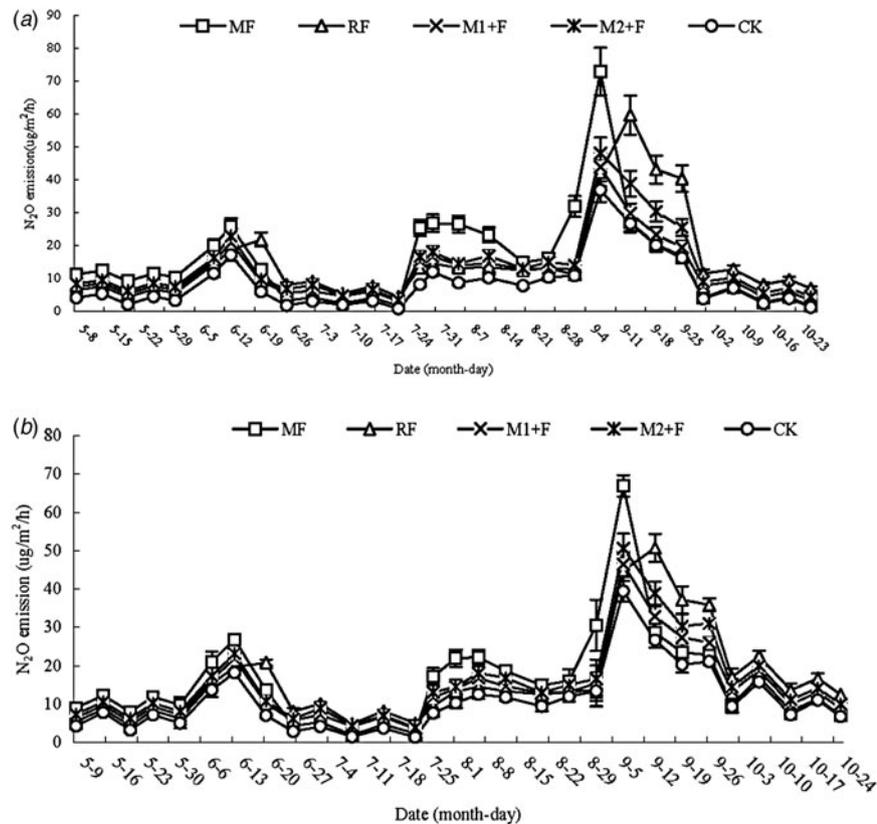
$0.007 \text{ g/m}^2$  (77.78%) in M2 + F in 2013, and by  $0.009 \text{ g/m}^2$  (81.82%) in MF,  $0.009 \text{ g/m}^2$  (81.82%) in RF,  $0.007 \text{ g/m}^2$  (63.64%) in M1 + F and  $0.008 \text{ g/m}^2$  (72.73%) in M2 + F in 2014 (Table 2). Similar results were observed in the late rice crop in both the seasons. Total  $\text{N}_2\text{O}$  emissions increased by  $0.053 \text{ g/m}^2$  (151.43%) in MF,  $0.052 \text{ g/m}^2$  (148.57%) in RF,  $0.039 \text{ g/m}^2$  (111.43%) in M1 + F and  $0.047 \text{ g/m}^2$  (134.29%) in M2 + F in 2013, and by  $0.055 \text{ g/m}^2$  (127.91%) in MF,  $0.055 \text{ g/m}^2$  (127.91%) in RF,  $0.047 \text{ g/m}^2$  (109.30%) in M1 + F and  $0.053 \text{ g/m}^2$  (123.26%) in M2 + F in 2014 (Table 2).

#### Global warming potentials of methane and nitrous oxide

Treatment M2 + F had larger total  $\text{CH}_4$  emissions than other treatments across the double rice cropping period, while MF and RF had the largest total  $\text{N}_2\text{O}$  emissions with quantities of  $0.062$  and  $0.061 \text{ g/m}^2$  in

2013, and  $0.064$  and  $0.064 \text{ g/m}^2$  in 2014, respectively (Tables 2 and 3).

Global warming potential reflects the relative effect of a GHG, with the GWP of  $\text{CO}_2$  being defined as 1. In the present study, the GWP of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  from double-cropping paddy fields varied with different fertilizer management, with the trend  $\text{M2 + F} > \text{M1 + F} > \text{RF} > \text{MF} > \text{CK}$ . In 2013, M2 + F had the largest GWP ( $3320.60 \text{ kg CO}_2\text{-equivalent/ha}$ ) of total  $\text{CH}_4$  and  $\text{N}_2\text{O}$  from double-cropping paddy fields, followed by M1 + F ( $2496.57 \text{ kg CO}_2\text{-equivalent/ha}$ ), RF ( $2124.63 \text{ kg CO}_2\text{-equivalent/ha}$ ) and CK ( $1257.93 \text{ kg CO}_2\text{-equivalent/ha}$ ). In 2014, M2 + F again had the largest GWP ( $2982.57 \text{ kg CO}_2\text{-equivalent/ha}$ ), followed by M1 + F ( $2521.55 \text{ kg CO}_2\text{-equivalent/ha}$ ), RF ( $2289.11 \text{ kg CO}_2\text{-equivalent/ha}$ ) and CK ( $1489.78 \text{ kg CO}_2\text{-equivalent/ha}$ ). According to GWP, the contribution of  $\text{CH}_4$  from double-cropping paddy fields to global warming was greater than that of  $\text{N}_2\text{O}$  (Table 3). Double rice grain yield was highest for RF and lowest for CK (Table 3).



**Fig. 5.** Nitrous oxide (N<sub>2</sub>O) fluxes from the soils affected by long-term fertilizer management during early and late cropped rice in (a) 2013 and (b) 2014. MF, mineral fertilizer alone; RF, crop residue + mineral fertilizer; M1 + F, low manure rate + mineral fertilizer; M2 + F, high manure rate + mineral fertilizer; CK, without fertilizer; ERT, early rice transplanting; ERH, early rice harvesting; LRT, late rice transplanting. Nitrous oxide emission rate is the mean of values measured within each treatment ( $n = 3$ ). Bars show s.d.

## DISCUSSION

### Methane emission

Methane emission is a complex process including production, oxidation and emission. Chu *et al.* (2007) reported that N fertilizer application decreased atmospheric CH<sub>4</sub> uptake and resulted in positive emissions from the soil. In the present study, the CH<sub>4</sub> flux and total CH<sub>4</sub> emissions from paddy fields during the early and late cropped rice were much larger in M2 + F, M1 + F and RF compared with CK, which was similar to the results of Wang *et al.* (2013). The reasons for this may be: (i) microbial activities were improved after returning crop residue/manure to the soil, due to the supplementation of carbon (C) sources and energy for microbial activities to accelerate consumption of soil oxygen and decrease soil redox potential (Eh); (ii) methanogens became active due to the large quantities of C sources, which provided reactive substrates for CH<sub>4</sub> emission from paddy fields. Meanwhile, higher CH<sub>4</sub> emission in

RF, M1 + F and M2 + F during the early and late cropped rice suggests higher root growth due to increased N supply by mineral fertilizer, manure or crop residue, which probably stimulated the activity of methanogens that produce CH<sub>4</sub>. When N was supplied with mineral fertilizer alone, as in the MF treatment, CH<sub>4</sub> emission was probably reduced because of the excessive soil inorganic N level. Reduced root growth due to lower levels of soil inorganic N as a result of the absence of fertilization with organic N probably also reduced methanotroph activity, thereby resulting in lower CH<sub>4</sub> emission in MF. Several researchers (Bronson & Mosier 1994; Powlson *et al.* 1997) have reported that inorganic N fertilization reduced soil CH<sub>4</sub> emission compared with organic N fertilization, while others (Amos *et al.* 2005; Mosier *et al.* 2006) did not observe the effects of N fertilization on emissions.

Methane production has been shown to increase with increasing temperature (Bergman *et al.* 1998), while CH<sub>4</sub> oxidation is less temperature-dependent

Table 2. Effects of long-term fertilizer managements on methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) emission from rice fields during whole growth period of early and late rice (g/m<sup>2</sup>)

Years	Treatment	CH <sub>4</sub>			N <sub>2</sub> O		
		Early rice	Late rice	Total	Early rice	Late rice	Total
2013	MF	2.30 ± 0.12*	3.40 ± 0.24	5.90 ± 0.36	0.017 ± 0.0011	0.044 ± 0.0014	0.062 ± 0.0023
	RF	3.50 ± 0.10	4.30 ± 0.17	7.80 ± 0.27	0.016 ± 0.0012	0.045 ± 0.0014	0.061 ± 0.0022
	M1 + F	3.50 ± 0.10	5.90 ± 0.12	9.40 ± 0.22	0.014 ± 0.0011	0.034 ± 0.0012	0.048 ± 0.0021
	M2 + F	4.17 ± 0.073	8.43 ± 0.097	12.60 ± 0.17	0.016 ± 0.0012	0.040 ± 0.0013	0.056 ± 0.0018
	CK	1.96 ± 0.057	2.65 ± 0.077	4.60 ± 0.13	0.009 ± 0.0009	0.026 ± 0.0011	0.035 ± 0.0014
2014	MF	3.50 ± 0.15	3.20 ± 0.18	6.70 ± 0.32	0.018 ± 0.0012	0.046 ± 0.0014	0.064 ± 0.0024
	RF	4.40 ± 0.13	4.00 ± 0.14	8.40 ± 0.27	0.018 ± 0.0012	0.047 ± 0.0014	0.064 ± 0.0024
	M1 + F	4.50 ± 0.13	4.91 ± 0.11	9.40 ± 0.24	0.016 ± 0.0011	0.040 ± 0.0012	0.056 ± 0.0023
	M2 + F	5.10 ± 0.10	6.10 ± 0.093	11.20 ± 0.19	0.017 ± 0.0011	0.045 ± 0.0013	0.062 ± 0.0025
	CK	2.89 ± 0.083	2.558 ± 0.074	5.40 ± 0.16	0.011 ± 0.0010	0.032 ± 0.0011	0.043 ± 0.0016

CK, without fertilizer; MF, mineral fertilizer alone; RF, crop residue plus mineral fertilizer; M1 + F, low manure rate plus mineral fertilizer; M2 + F, high manure rate plus mineral fertilizer.

\* Values are presented as mean ± S.E. (*n* = 3).

than CH<sub>4</sub> production (Dunfield *et al.* 1993). The higher CH<sub>4</sub> emissions in all treatments may result from higher CH<sub>4</sub> production than the consumption rate under the elevated soil temperature, and could partially explain why CH<sub>4</sub> emissions increased in late rice growth season than that of the early rice growth season. In the present study, CH<sub>4</sub> emissions increased during the early and late cropped rice when soil Eh decreased, and there are differences in CH<sub>4</sub> emissions observed among the different fertilizer management practices. As soil Eh increased during the early and late cropped rice, CH<sub>4</sub> emissions decreased.

Hu *et al.* (2002) similarly observed significant CH<sub>4</sub> emissions in paddy fields and significantly higher CH<sub>4</sub> emissions with N fertilization. In the present study, CH<sub>4</sub> fluxes from the control were low in the early and late cropped rice. When N was supplied jointly by manure, crop residue and mineral fertilizer, such as in M2 + F with high manure rate plus mineral fertilizer, M1 + F with low manure rate plus mineral fertilizer and RF with crop residue plus mineral fertilizer, CH<sub>4</sub> emissions were probably increased because of the high levels of soil organic N. Therefore, during the early and late cropped rice, the CH<sub>4</sub> emission increased gradually with the decomposition of organic matters and growth of rice after transplanting, and reached the peak value at tillering stage in all treatments. However, CH<sub>4</sub> emissions in both rice seasons were reduced to a large extent after field drying, because soil aeration was improved during this period, and the activities of methanogens were

therefore restricted, and the physiological activity of rice plants decreased, thereby limiting the ability for transportation and emission of CH<sub>4</sub>. Compared with the RF, M1 + F and M2 + F, it was also observed that MF decreased CH<sub>4</sub> emissions from the paddy soil. These results indicated that in paddy soils, where there is high precipitation and high soil temperature, CH<sub>4</sub> production may occur.

#### Nitrous oxide emission

Nitrous oxide is emitted by soils as a result of denitrification in anaerobic soil and nitrification in aerobic soil, with the anaerobic production considered to be more important. In the present study, N<sub>2</sub>O fluxes from the control were lower than those from the organic–inorganic mixed mineral fertilizer treatments throughout the whole experimental period. The greater N<sub>2</sub>O flux in MF and RF than in the control during early and late cropped rice, however, probably resulted from increased N substrate availability from both crop residue and mineral fertilizer. Increased N substrate availability due to N fertilization has been known to increase N<sub>2</sub>O flux due to enhanced nitrification (Drury *et al.* 2006; Mosier *et al.* 2006; Dusenbury *et al.* 2008). Increased emissions of N<sub>2</sub>O from paddy fields by organic–inorganic mixed fertilization management practices have also been reported in other studies (Akiyama *et al.* 2000; Yan *et al.* 2001; Akiyama & Tsuruta 2002; Li *et al.* 2002; Chu *et al.* 2007). In the present study, organic–inorganic mixed

Table 3. Double rice grain yield, global warming potentials (GWP) of methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) and per yield GWP CO<sub>2</sub> from rice fields under long-term fertilizer managements

Years	Treatment	CH <sub>4</sub> emission (g/m <sup>2</sup> )	N <sub>2</sub> O emission (g/m <sup>2</sup> )	GWP of CH <sub>4</sub> (kg CO <sub>2</sub> /ha)	GWP of N <sub>2</sub> O (kg CO <sub>2</sub> /ha)	GWP of CH <sub>4</sub> and N <sub>2</sub> O (kg CO <sub>2</sub> /ha)	Early and late rice grain yield (kg/ha)	Per yield GWP CO <sub>2</sub> (kg/kg)
2013	MF	5.90 ± 0.36*	0.06 ± 0.002	1470 ± 91	185 ± 5.3	1655 ± 96	10364 ± 312.5	0.16 ± 0.007
	RF	7.80 ± 0.27	0.06 ± 0.002	1942 ± 68	183 ± 5.3	2125 ± 72	11473 ± 299.2	0.19 ± 0.009
	M1 + F	9.40 ± 0.22	0.05 ± 0.002	2352 ± 56	145 ± 4.8	2497 ± 61	10691 ± 308.6	0.23 ± 0.007
	M2 + F	12.60 ± 0.17	0.06 ± 0.001	3154 ± 43	166 ± 4.2	3321 ± 48	10824 ± 331.2	0.31 ± 0.005
2014	CK	4.60 ± 0.13	0.04 ± 0.001	1155 ± 33	103 ± 3.0	1258 ± 36	5069 ± 146.3	0.25 ± 0.006
	MF	6.70 ± 0.32	0.06 ± 0.002	1673 ± 81	192 ± 5.5	1865 ± 86	10480 ± 318.7	0.18 ± 0.011
	RF	8.40 ± 0.27	0.06 ± 0.002	2098 ± 68	191 ± 5.5	2289 ± 73	12098 ± 302.5	0.19 ± 0.012
	M1 + F	9.40 ± 0.24	0.06 ± 0.002	2354 ± 61	168 ± 5.4	2522 ± 66	10936 ± 315.7	0.23 ± 0.015
	M2 + F	11.20 ± 0.19	0.06 ± 0.002	2797 ± 48	186 ± 4.9	2983 ± 54	11039 ± 349.2	0.27 ± 0.016
	CK	5.4 ± 0.16	0.043 ± 0.001	1361 ± 39.3	129 ± 3.7	1490 ± 43.0	5723 ± 165.2	0.26 ± 0.015

CK, without fertilizer; MF, mineral fertilizer alone; RF, crop residue plus mineral fertilizer; M1 + F, low manure rate plus mineral fertilizer; M2 + F, high manure rate plus mineral fertilizer.

\* Values are presented as mean ± s.e. (n = 3).

fertilization management practices increased emissions of N<sub>2</sub>O during the early and late cropped rice. The greater N<sub>2</sub>O flux in M2 + F and M1 + F was probably due to both the organic and the inorganic mineral fertilization and the flux in early and late cropped rice was probably a result of substantial precipitation and/or increased soil temperature. It has been suggested that the accumulation of manure and crop residue material in the soil during growing of early and late rice enhanced N<sub>2</sub>O production (Christensen & Christensen 1991).

Several researchers have noted increased N<sub>2</sub>O flux immediately after N fertilization and/or substantial precipitation (Mosier *et al.* 2006; Dusenbury *et al.* 2008; Liebig *et al.* 2010). In the present study, greater N<sub>2</sub>O flux in MF than in M2 + F, M1 + F and RF during the rice growing season may have resulted from increased N contribution from mineral fertilizer, due to its higher N concentration. Greater N<sub>2</sub>O flux in M2 + F, M1 + F and RF than in the control during the early and late cropped rice may have resulted either from increased N contribution from crop residue due to the higher N concentration in RF and increased organic N mineralization due to manure application during rice growing season in M2 + F and M1 + F. The reasons for greater N<sub>2</sub>O flux in M2 + F, M1 + F and RF in late rice growth period were also likely to be a result of increased microbial activity and soil temperature. Meanwhile, compared with M1 + F, the greater N<sub>2</sub>O flux in M2 + F with high manure rate plus mineral fertilizer probably resulted from increased N substrate availability from both high manure and mineral fertilizer. Increased N substrate availability due to N fertilization has been confirmed to increase N<sub>2</sub>O flux due to enhanced nitrification (Drury *et al.* 2006; Mosier *et al.* 2006; Dusenbury *et al.* 2008).

#### Effects of methane and nitrous oxide flux factors

A considerable number of studies have shown that some soil or environmental factors can influence CH<sub>4</sub> and N<sub>2</sub>O emission. For example, CH<sub>4</sub> and N<sub>2</sub>O production is regulated by vegetation type, soil temperature, soil moisture, root activity and many other factors (Parkin & Kaspar 2003; Wassmann *et al.* 2004; Ma *et al.* 2008; Kallenbach *et al.* 2010), and soil temperature, soil moisture and soil Eh have been determined to be the most crucial regulators (Kudo *et al.* 2014). Yu *et al.* (2007) reported that CH<sub>4</sub> emission showed an exponential decrease when Eh

increased. In the present study, during the early and late cropped rice, the soil Eh rapidly increased simultaneously with rapid CH<sub>4</sub> emission decrease. In the present study, crop residues and manure were incorporated into the soil in the RF, M1 + F, M2 + F treatments and their decomposition consumed limited soil-dissolved oxygen. All these factors resulted in decreased Eh and consequently increased CH<sub>4</sub> emission under RF, M1 + F and M2 + F.

The soil temperature had a predictive functional relationship with CH<sub>4</sub> emission. Khalil *et al.* (1998) observed an increase in CH<sub>4</sub> emissions from paddy fields with increasing soil temperature and Zhu *et al.* (2007) reported a strong correlation between CH<sub>4</sub> emission and soil temperature. The current results also found that there was a relationship between CH<sub>4</sub> emission and soil temperature, with soil temperature found to be a major factor affecting CH<sub>4</sub> emission. In general MF decreased soil temperature, especially during the hotter days, and this may have been partly responsible for lower CH<sub>4</sub> emissions when compared with other treatments. Temperature was also the major reason for differences in the CH<sub>4</sub> emission pattern between the early and late cropped rice. In the current experimental area, the late rice season was the hottest time of the summer and high temperatures enhanced the decomposition rate of crop residues and manure in the moist environment. During decomposition, a large number of organic compounds are produced and oxygen is consumed, thus decreasing soil Eh and leading to increased CH<sub>4</sub> emission. In contrast to the warm temperatures of the late rice season, air temperatures in the early rice season were lower, which resulted in slower crop residue and manure decomposition and therefore little CH<sub>4</sub>-substrate. Hence, these differences in weather factors (e.g. temperature) resulted in the different characteristics of CH<sub>4</sub> between the early and late cropped rice.

The N<sub>2</sub>O emission was influenced strongly by external factors and many emission peaks occurred during the rice growing season. The emission of N<sub>2</sub>O was different between the early and late cropped rice, possibly due to the variations in weather. Some studies show that extreme precipitation and drying could increase N<sub>2</sub>O emission (Zona *et al.* 2011). Hao *et al.* (2001) reported that aeration and water flooding led to 'outbreaks' of emissions. In the present study, precipitation in the early rice growing season was much higher than that in the late season. This precipitation difference may explain the fluctuations of N<sub>2</sub>O emissions between the seasons.

In addition, similar to CH<sub>4</sub>, N<sub>2</sub>O emission is also influenced by soil Eh. Weier *et al.* (1993) reported that the rate of N<sub>2</sub>O emission decreased with increasing soil reducibility. In the present study, crop residues and manure in RF, M1 + F, M2 + F were mainly distributed within the plough layer (0–20 cm) and had a strong redox potential due to decomposition of crop residues and manure. Therefore, compared with the MF, N<sub>2</sub>O produced from RF, M1 + F, M2 + F soils tended to be further deoxidized to nitrogen gas (N<sub>2</sub>), which consequently decreased N<sub>2</sub>O emission. Meanwhile, the greater N<sub>2</sub>O flux in the late rice growing season than in the season was probably due to increased soil temperature, since increased temperature can stimulate microbial activity and N mineralization (Parkin & Kaspar 2003; Dusenbury *et al.* 2008; Liebig *et al.* 2010).

#### Global warming potentials of methane and nitrous oxide

Global warming potential can be used as an index to estimate the potential effects of different GHGs on the global climate system. Tang *et al.* (2014) estimated that GWP of double-cropping rice systems increased through the return of straw from winter cover crops. Zhu *et al.* (2012) reported that the highest GWP was found for incorporation of Chinese milk vetch in a double-cropping rice system, which was 21–325% higher than the other treatments they studied. In the present study, the GWP of CH<sub>4</sub>, N<sub>2</sub>O or both had different orders. For a comprehensive consideration, GWP of both CH<sub>4</sub> and N<sub>2</sub>O is an important method to assess the effect of a farming system on climate warming. Therefore, it is necessary to make a combined estimate of global warming effects of CH<sub>4</sub> and N<sub>2</sub>O emitted from each treatment. Thus, GWP and GWP per yield were introduced into the present study for global warming calculations. It was found that the CH<sub>4</sub> and N<sub>2</sub>O GWP for M2 + F and M1 + F were higher than for RF and MF, due to their greater CH<sub>4</sub> emissions. Manure and crop residue addition increased early and late rice grain production compared with the control. Therefore, the control without fertilizer may mitigate GHG emissions but may not sustain rice yields. The total of early and late rice production was significantly higher in the M2 + F, M1 + F and RF than in the control, but the GWP per yield of RF was significantly lower than the control, M2 + F and M1 + F. Therefore, application of the crop residue and mineral fertilizer pattern is

recommended for double-cropping rice areas in the Middle and Lower reaches of Yangtze River in China, which corresponds to RF as a management option under an intensive cropping system. Further studies are under way to examine whether RF with reduced N fertilization rate might mitigate GHG emissions and sustain crop yields. Other benefits of applying crop rotation rather than mono-cropping include reduced infestation of weeds, diseases and pests. However, for evaluating the GWP of management systems, it would be mandatory to consider the soil C dynamics associated with crop production inputs and machinery use.

## CONCLUSIONS

Greenhouse gas emissions from large paddy fields and excessive fertilizer application can contribute significantly to global warming. Managing fertilizer application is one of the feasible ways of limiting GHG emissions from paddy areas. Low fertilizer application results in low-energy consumption, which can contribute to the reduction of GHG and lessen global warming. The current results show that mineral fertilizer application stimulates N<sub>2</sub>O emission during the early and late cropped rice. Meanwhile, the results indicate that with the same N application rate, different organic–inorganic mixed fertilizer application, such as RF, M1 + F and M2 + F, caused substantial CH<sub>4</sub> emissions during the early and late cropped rice compared with those from the conventional MF treatment. However, it significantly increased rice grain yields in both RF, M1 + F and M2 + F production systems. The GWP of both CH<sub>4</sub> and N<sub>2</sub>O resulted in a significantly lower yield-scaled GWP compared with the control treatments. The increased use of organic inputs greatly affected CH<sub>4</sub> and N<sub>2</sub>O emission; however, the inputs were necessary for maintaining soil fertility and are a crucial source of nutrient input for small-scale farmers in paddy fields who cannot rely solely on mineral fertilizer. The combined use of organic inputs and mineral fertilizer is therefore suggested as a potential for intensifying rice production in paddy soil under intensive cropping systems. Nevertheless, future research is necessary to provide a complete insight into the effects of recently developed practices on soil C dynamics.

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