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EFFECTS OF FERTILIZATION ON POREWATER NUTRIENTS, GREENHOUSE-GAS EMISSIONS AND RICE PRODUCTIVITY IN A SUBTROPICAL PADDY FIELD

By WEIQI WANG^{1,2*}, JORDI SARDANS^{3,4*}, CHUN WANG^{1,2}, CHUAN TONG^{1,2}, QINYANG JI^{1,2}, JOSEP PEÑUELAS^{3,4}

[1]Institute of Geography, Fujian Normal University, Fuzhou 350007, China
[2]Key Laboratory of Humid Subtropical Eco-geographical Process, Ministry of Education, Fujian Normal University, Fuzhou 350007, China
[3]CSIC, Global Ecology CREAF- CSIC-UAB, Cerdanyola del Valles, 08193 Barcelona, Catalonia, Spain
[4] CREAF, Cerdanyola del Valles, 08193 Barcelona, Catalonia, Spain

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Corresponding author:
Weiqi Wang. Email: wangweiqi15@163.com; Phone: +86-591-83465214;
Fax: +86-591-83465397

SUMMARY

Suitable fertilization is crucial for the sustainability of rice production and for the potential mitigation of global warming. The effects of fertilization on porewater nutrients and greenhouse-gas fluxes in cropland, however, remain poorly known. We studied the effects of no fertilization (control), standard fertilization and double fertilization on the concentrations of porewater nutrients, greenhouse-gas fluxes and emissions and rice yield in a subtropical paddy in southeastern China. Double fertilization increased dissolved NH_4^+ in porewater. Mean CO_2 and CH_4 emissions were 13.5% and 7.4%, 20.4% and 39.5% higher for the standard and double fertilizations than the control, respectively. N_2O depositions in soils were 61% and 101% greater for the standard and double fertilizations than the control, respectively. The total global warming potentials (GWPs) for all emissions were 14.1% and 10.8% higher for the standard and double fertilizations than the control, respectively, with increasing contribution of CH_4 with fertilization and a CO_2 contribution > 85%. The total GWPs per unit yield were significantly higher in the standard and double fertilizations than the control by 7.3% and 10.9%, respectively. The two levels of fertilization did not significantly increase rice yield. Prior long-term fertilization in the paddy (about 20 years with annual doses of 95 kg N ha^{-1}, 70 kg P_2O_5 ha^{-1} and 70 kg K_2O ha^{-1}) may have prevented these fertilizations from increasing the yield. However, fertilizations increased greenhouse-gas emissions. This situation is common in paddy fields in subtropical China, suggesting a saturation of soil nutrients and the necessity to review current fertilization management. These areas likely suffer from unnecessary nutrient leaching and excessive greenhouse-gas emissions. These results provide a scientific basis for continued research to identify an easy and optimal

fertilization management.

Keywords: Paddy field; CH_4 flux; N_2O flux; CO_2 flux; porewater nutrient; fertilizer

INTRODUCTION

Greenhouse gases (GHGs), such as carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O), contribute about 80% to the current global radiative forcing (Myhre et al., 2013). Agricultural activities contribute approximately 20% of the present concentrations of atmospheric GHGs (Hütsch, 2001), especially the emissions of CH_4 and N_2O from paddy fields (Myhre et al., 2013), so minimizing the release of these GHGs from paddies and thus mitigating their adverse impacts on climate change are of the utmost importance. As a main cereal crop, rice currently feeds more than 50% of the global population (Haque et al., 2015) and rice production will need to increase by 40% by the end of 2030 to meet the demand for food from the growing population worldwide (FAO, 2009).

Fertilization is important for sustainable rice production (Linquist et al., 2013) and numerous studies have been devoted to the development of suitable practices of fertilizer management for both improving rice yields and mitigating GHG emissions, including the application of fertilizers such as straw mulch (Dossou-Yovo et al., 2016), silicate fertilizer (Wang et al., 2015) and the establishment the most adequate N, P and K fertilizers (Thao et al., 2015). Fertilizers are key for rice productivity, but the amounts needed for maintaining rice yield and minimizing environmental effects are unknown. Chemical fertilizers are necessary to keep the world's population fed, but their overuse threatens the safety of the soil, water and air (Zhang et al., 2013a). China has become the world's largest consumer of fertilizer, and its fertilizer-use efficiency is much less than half the amendment amount (Cheng and Li, 2007). Determining the amount suitable for supporting rice productivity and reducing nutrient losses is therefore very important (Zhao et al., 2015).

Chemical fertilizers, especially N fertilizers, influence the dynamics of GHGs the most (Zhong et al., 2016). Zhao et al. (2015) reported that N_2O emissions increased but CH_4 emissions decreased with the level of fertilization. Zhong et al. (2016) indicated that the global warming potential (GWP) and rice yields increased with the rate of application of nitrogen fertilizer. In contrast, Zhang et al. (2013a) showed that N fertilization could reduce GHG emissions. Some studies have showed that maintaining soil fertility and crop productivity and at the same time reducing GHGs emissions have several trade off questions to take into account (Bhatia et al., 2005). The substitution of inorganic N fertilizers by organic crop manure residues improves yield and soil health but can increase CH_4 emissions (Bhatia et al., 2005; 2012). At medium-long term, the substitution of inorganic by organic residues as fertilizers improve soil aggregate stability, soil water holding capacity and soil microbial activity (Sihi et al, 2016a; 2017) without any serious decrease in yield production (Sihi et al., 2012). Thus, establishing suitable fertilization rates to ensure rice yields and reduce GHG emissions is important for field management (Zhong et al., 2016). The control of water inundation management and the time to plant and sowing rice plants (Dari et al., 2017) or the use of crop manure and/or urea plus dicyanamide have reduced N_2O emissions in paddy soils without reducing yield (Pathak et al., 2002) and increased the recovery efficiency of N added by fertilizers (Pathak 2010). Anyway, fertilization management influences soil properties and this affects GHGs emissions in wetland areas (Davidson and Janssens, 2006), and there is not a general consensus in the adequate fertilization management for an equilibrated soil fertility and rice production without increased GHGs emissions.

China has the world's second largest area of rice cultivation, and the associated GHG emissions account for about 40% of the total agricultural GHG emission. In China, 90% of paddies are in the subtropics, such as in Fujian, Jiangxi and Hunan Provinces. Developing effective strategies to increase the cost-effectiveness of rice agriculture, enhancing crop yield and mitigating GHG emissions from paddies in subtropical China to minimize future food shortages and adverse climate change is thus a global objective of national importance. We pursued this objective by: (1) determining the emissions of CO_2, CH_4 and N_2O in response to the application of different amounts of fertilizers in paddy fields; (2) exploring the effect of amendment amount on the concentrations of porewater nutrients; and (3) assessing the impacts of the fertilizer applications on crop productivity. The results obtained in this study will provide a scientific basis for the suitable management of fertilization for rice agriculture and the evaluation of most current fertilization strategies for increasing yield and decreasing GHG emissions.

MATERIALS AND METHODS

Study site

Our study was conducted at the Wufeng Agronomy Field of the Fujian Academy of Agricultural Sciences in Fujian Province, southeastern China (26.1°N, 119.3°E) (Supplementary Material Fig. S1). A field experiment was carried out during the early paddy season (16 April to 16 July) in 2014. Air temperature and humidity during the study period are shown in Fig. S2. The soil of the paddy was poorly drained, and the proportions of sand, silt and clay particles in the top 15 cm of the soil were 28%, 60% and 12%, respectively (Wang et al., 2015). Other properties of the top 15 cm of soil at the beginning of the experiment were: bulk density, 1.1 g cm^{-3}; pH (1:5 with H_2O), 6.5; organic carbon (C) content, 18.1 g kg^{-1}; total nitrogen (TN) content, 1.2 g kg^{-1} and total phosphorus (TP) content, 1.1 g kg^{-1} (Wang et al., 2015). Water level was maintained at 5-7 cm above the soil surface throughout the growing season by an automatic water-level controller, and the paddy was last drained two weeks before harvest.

We established triplicate plots (10 × 10 m) for two treatments and a control in which rice seedlings (cultivar Hesheng 10) were inserted to a depth of 5 cm with a spacing of 14 × 28 cm using a rice transplanter. The age of the rice seedlings was 21 days when they were transplanted. The two treatments consisted of standard fertilization and fertilization with double amount of the standard fertilization. The control plots were not fertilized. The standard fertilization treatment consisted of three applications of N-P_2O_5-K_2O (16-16-16%; Keda Fertilizer Co., Ltd. Jingzhou, China) and urea (46% N) fertilizers. The first application was one day before transplantation at rates of 42 kg N ha^{-1}, 40 kg P_2O_5 ha^{-1} and 40 kg K_2O ha^{-1}. The second application was broadcasted during tiller initiation (seven days after transplanting (DAT)) at rates of 35 kg N ha^{-1}, 20 kg P_2O_5 ha^{-1} and 20 kg K_2O ha^{-1}. The third application was broadcasted during panicle initiation (56 DAT) at rates of 18 kg N ha^{-1}, 10 kg P_2O_5 ha^{-1} and 10 kg K_2O ha^{-1}. These doses are the most commonly used in paddy fields in subtropical China (with annual doses of 95 kg N ha^{-1}, 70 kg P_2O_5 ha^{-1} and 70 kg K_2O ha^{-1}), which constitute about 90% of the paddy fields in China (Wang et al., 2015). The double-fertilization treatment consisted of twice the amounts of the standard fertilization but the same schedule. The plots had previously been fertilized with amounts equal to the standard fertilization during a period of about 20 years (Wang et al., 2015). All control and treatment plots received the same amount of water. The field was plowed to a depth of 15 cm with a moldboard plow and was leveled two days before rice transplantation.

Measurement of CO_2, CH_4 and N_2O emissions

Static closed chambers were used to measure CO_2, CH_4 and N_2O emissions. The chambers were made of PVC and consisted of two parts, an upper transparent compartment (100 cm high, 30 cm wide, 30 cm long) placed on a permanently installed bottom collar (10 cm high, 30 cm wide, 30 cm long). Each chamber had two battery-operated fans to mix the air inside the chamber headspace, an internal thermometer to monitor temperature changes during gas sampling and a gas-sampling port with a neoprene rubber septum at the top of the chamber for collecting gas samples from the headspace. We deployed three replicate chambers in each plot. A wooden boardwalk was built for accessing the plots to minimize the disturbance of the soil during gas sampling. The chambers were installed on soil with plants inside..

Gas flux was measured weekly in all chambers. Gas samples were collected from the chamber headspace using a 100-mL plastic syringe with a three-way stopcock 0, 15 and 30 min after deployment of the upper compartment. The samples were immediately transferred to 100-mL air-evacuated aluminum foil bags (Delin Gas Packaging Co., Ltd., Dalian, China) sealed with butyl rubber septa and transported to the laboratory for the analysis of CO_2, CH_4 and N_2O.

CO_2, CH_4 and N_2O concentrations in the headspace air samples were determined by gas chromatography (Shimadzu GC-2010 and Shimadzu GC-2014, Kyoto, Japan) using a stainless steel Porapak Q column (2 m long, 4 mm outer diameter, 80/100 mesh). A methane conversion furnace, flame ionization detector (FID) and electron capture detector (ECD) were used for the determination of the CO_2, CH_4 and N_2O concentrations, respectively. The operating temperatures of the column, injector and detector for the determination of CO_2, CH_4 and N_2O concentrations were adjusted to 45, 100 and 280 °C; to 70, 200 and 200 °C and to 70, 200 and 320 °C, respectively. These temperatures were the optimum temperatures for the different parts of the instrument. Helium (99.999% purity) was used as a carrier gas (30 mL min^{-1}), and a make-up gas (95% argon and 5% CH_4) was used for the ECD. The gas chromatograph was calibrated before and after each set of measurements using 503, 1030 and 2980 μL CO_2 L^{-1} in He; 1.01, 7.99 and 50.5 μL CH_4 L^{-1} in He and 0.2, 0.6 and 1.0 μL N_2O L^{-1} in He (CRM/RM Information Center of China) as primary standards. CO_2, CH_4 and N_2O fluxes were then calculated as the rate of change in the mass of CO_2, CH_4 and N_2O per unit of surface area and per unit of time by using a closed-chamber equation (Ali et al., 2008):

F= ((M/V) x (dc/dt)) x H x (273/(273 + T)), where F is the corresponding gas flux (mg/mg m^{-2} h^{-1}), M is the molecular weight, V is the height of the chamber above water surface (m), and T is the air temperature inside the chamber (ºC).

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Global warming potential (GWP)

GWP is typically estimated using CO_2 as the reference gas, and a change in the emission of CH_4 or N_2O is converted into "CO_2-equivalents" (Hou et al., 2012). The GWP for CH_4 is 34 (based on a 100-year time horizon and a GWP for CO_2 of 1), and the GWP for N_2O is 298 (Myhre et al., 2013). The GWP of the combined emission of CH_4 and N_2O was calculated as (Ahmad et al., 2009): GWP = (cumulative CO_2 emission × 1 + cumulative CH_4 emission × 34 + cumulative N_2O emission × 298).

Collection of soil porewater

Porewater was sampled *in situ* once a week from 16 April to 16 July 2014. Three specially designed PVC tubes (5.0 cm inner diameter) were installed to a depth of 15 cm in each plot. Porewater samples were collected using 50-mL syringes and then separated into two parts: about 10 mL were injected into pre-evacuated 20-mL vials, and the remaining 40 mL were injected into 100-mL sample vials. The samples were stored in a cool insulated box in the field until transported to the laboratory where they were stored at -20 °C until the analysis of nutrients and GHG concentrations.

Measurement and calculation of dissolved CO_2, CH_4 and N_2O concentrations

The sample vials were thawed at room temperature and then vigorously shaken for 5 min to equilibrate the CH_4 concentrations between the water and the headspace. Gas samples were collected from the headspaces of the vials and analyzed for CO_2, CH_4 and N_2O concentrations by gas chromatography (Shimadzu GC-2010 and Shimadzu GC-2014, Kyoto, Japan; see above for more details).

The concentrations (C) of CO_2, CH_4 and N_2O dissolved in the water were calculated following Ding et al. (2003): C = (Ch × Vh)/(22.4 × Vp), where Ch is the CO_2, CH_4 and N_2O concentration (μL L^{-1}) in the air samples from the vials, Vh is the volume of air in the vial (mL) and Vp is the volume of the water in the vial (mL).

Measurement of soil, porewater properties and rice yield

Soil temperature, pH, salinity, redox potential (Eh) and water content of the top 15 cm of soil were measured in triplicate in situ in each plot on each sampling day. Temperature, pH and Eh were measured with an Eh/pH/Temperature meter (IQ Scientific Instruments, Carlsbad, USA), salinity was measured using a 2265FS EC meter (Spectrum Technologies Inc., Paxinos, USA) and water content was measured using a TDR300 meter (Spectrum Field Scout Inc., Aurora, USA). The concentrations of NH_4^+, NO_3^-, TN and TP in the porewater were determined using a sequence flow analyzer (San^{++}, SKALAR Corporation production, Breda, The Netherlands). The concentration of dissolved organic carbon (DOC) was determined using a TOC Analyzer (TOC-V CPH, Shimadzu Corporation, Kyoto, Japan) and a filter paper of 60 μm of pore diameter. Rice yield was determined at the harvesting stage by manual harvest (Wang et al., 2015).

Statistical analysis

Differences in soil properties; CO_2, CH_4 and N_2O emissions; dissolved porewater CO_2, CH_4 and N_2O concentrations and porewater nutrient concentrations among the treatments and control were tested for statistical significance by repeated-measures analyses of variance. The relationships between mean GHG emissions and the soil properties, dissolved porewater GHG concentrations and porewater nutrient concentrations were determined by Pearson correlation analysis. These statistical analyses were performed using SPSS Statistics 18.0 (SPSS Inc., Chicago, USA). We analyzed the effects of multiple soil variables as fixed factors on the production rates of the three GHGs using general linear models with and without spatial correlation. We used linear (lm) and mixed (lme) functions with the “nlme” and “lme4” R packages. We chose the best model for each dependent variable using the Akaike information criterion. We used the MuMIn R package in mixed models to estimate the percentage of variance explained by the model.

RESULTS

GHGs dissolved in porewater and emitted from the paddy

CO_2, CH_4 and N_2O emissions varied significantly among most sampling dates ($P<0.01$, Table S1) but the treatments and the interaction of sampling date and treatment were not significant ($P>0.05$). CO_2 flux generally remained low (< 354 mg m^{-2} h^{-1}) during the first 29 DAT but then increased to a seasonal peak (> 2811 mg m^{-2} h^{-1}) between 29 and 71 DAT (Fig. 1A). The rice was nearly ripe by 71 DAT, with a corresponding decrease in CO_2 emissions until harvesting in July. The CH_4 emissions were low soon after rice transplantation and peaked by 71 DAT in all treatments (Fig. 1B). The paddy was drained after the rice reached maturity, with a corresponding decrease in CH_4 emissions until harvesting in July.

Mean CO_2 emissions were 13.5% and 7.4% higher for the standard and double fertilizations than the control, respectively. Mean CH_4 emissions were 20.4% and 39.5% higher for the standard and double fertilizations than the control, respectively. Mean N_2O soil depositions were 61% and 101% higher for the standard and double fertilizations than the control, respectively, mostly due to the lower (negative) values at 36 DAT (Fig. 1C), despite no overall effect of the treatments on N_2O emission determined by the mixed linear models (Table 1). Dissolved CO_2, CH_4 and N_2O concentrations varied significantly among sampling dates ($P<0.01$, Table S1). Treatments and the interaction of date and treatment were significant for dissolved CO_2 concentration but not for dissolved CH_4 and N_2O concentrations ($P>0.05$; Table S1, Fig. 2).

Mean dissolved CO_2 concentration was 11.4% lower for the standard fertilization and 95.0% higher for the double fertilization than the control. Mean dissolved CH_4 concentrations were 25.8% and 18.9% lower for the standard and double fertilizations than the control, respectively. Mean dissolved N_2O concentrations were 21.0% and 73.5% higher for the standard and double fertilizations than the control, respectively.

The mixed linear models (with plot and time as random factors) showed that standard fertilization increased CO_2 emissions and soil Eh and salinity, whereas double fertilization increased CH_4 emission, porewater CO_2 concentration, soil salinity and porewater NH_4^+ and DOC concentrations (Table 1). This mixed linear model analysis is more robust to detect the effects of treatments than the previously commented ANOVA. CO_2 emission was correlated positively with soil Eh, temperature, water content and porewater NH_4^+ concentration and negatively with soil pH and porewater TN, TP and DOC concentrations (Table S2).

Soil and porewater properties of the paddy

Soil Eh, salinity and porewater NH_4^+, TP and DOC concentrations, soil pH, temperature, water content and porewater NO_3^- and TN concentrations varied significantly among sampling dates ($P<0.01$; Table S3, Figs. 3 and 4). The interaction of sampling date and treatment had significant effects on soil pH, temperature, water content and porewater NO_3^- and TN concentrations ($P<0.05$). Mean soil pH, Eh, salinity, water content and temperature for the standard and double fertilizations differed by <10% from the control. Mean porewater NH_4^+ concentrations were 114.8% and 213.7% higher for the standard and double fertilizations than the control, respectively. Mean porewater NO_3^- concentrations were 17.8% and 30.9% higher for the standard and double fertilizations than the control, respectively. Mean porewater TN concentrations were 19.7% and 42.0% higher for the standard and double fertilizations than the control, respectively. Mean porewater TP concentrations were 45.7% and 213.3% higher for the standard and double fertilizations than the control, respectively. Finally, mean porewater DOC concentrations were 9.3% and 11.7% higher for the standard and double fertilizations than the control, respectively.

Relationships among GHG emissions, dissolved GHG concentrations and soil and porewater properties

The mixed linear models (with plot and time as random factors) showed that CH_4 emissions were positively correlated with soil water content (Table S2). N_2O emission was correlated positively with porewater total TN concentration and negatively with soil pH. Porewater CO_2 concentrations were correlated positively with soil salinity and water content and negatively with porewater TN, TP, NH_4^+ and DOC concentrations. Porewater CH_4 concentrations were positively correlated with soil water content, and porewater N_2O concentrations were positively correlated with soil temperature and porewater NH_4^+ concentrations.

Soil relationships varied between the treatments (Tables S4 and S5). Seasonal CO_2 emission was positively correlated with soil Eh ($P<0.05$, Table 2) and temperature ($P<0.01$, Table S4) in all plots. Seasonal CH_4 emission was positively correlated with soil salinity ($P<0.01$) and water content ($P<0.05$) in all plots.

Dissolved CO_2 concentration was positively correlated with soil water content ($P<0.05$), while dissolved N_2O concentration was negatively correlated with soil temperature ($P<0.05$, Table S4) in all plots.

Seasonal CO_2 emission was positively correlated with CH_4 emission ($P<0.05$, Table S5) and dissolved CO_2 concentration ($P<0.01$). Seasonal CH_4 emission was positively correlated with dissolved CH_4 concentration ($P<0.01$) and dissolved CO_2 concentration was positively correlated with dissolved CH_4 concentration ($P<0.01$).

Rice productivity and GWP

The average rice yield was about 5.2% higher for the standard fertilization and about 1.4% lower for the double fertilization than the control, which were not significantly different (Table 2). Contribution of CO_2 to total GWP (> 85%) was higher than CH_4 and N_2O. The total GWPs for all emissions were 14.1% and 10.8% higher for the standard and double fertilizations than the control, respectively. The total GWPs per unit yield were significantly higher by 7.3% and 10.9% for the standard and double fertilizations than the control, respectively.

DISCUSSION

Effects of fertilization on CO_2 emissions

Mean CO_2 emissions were higher for the standard and double fertilizations than the control, for several potential reasons. First, fertilization, such as N fertilization, promotes the deposition of photosynthetically derived C into soil organic carbon pools. Then, soil respiration increases when inputs of active C substrates increase (Ge et al., 2015). Second, fertilizer can provide many nutrients for microbial growth (Inselsbacher et al., 2011), and the increase in microbial activity promotes soil respiration and thus CO_2 emission (Adewopo et al., 2015). Third, NH_4^+ from fertilizers can be oxidized to NO_3^- when paddies are drained, increasing soil NO_3^- concentration. This NO_3^- would be reduced when the paddies are reflooded, producing CO_2 (Wang et al., 2015). Moreover, NH_4^+ amendment in our study would be associated with ferric reduction, increasing the production and release of CO_2 (Luo et al., 2016). Ferric reduction should also decrease the number of iron plaques (by the higher solubility of Fe^{2+} than Fe^{3+}) on the rice roots, which would increase the transport of gases throughout the rice plants (Huang et al., 2012). Transport by rice plants is the most important pathway of gas emission to the atmosphere (Wassmann and Aulak, 2000). Decreases in the number of iron plaques will promote root ventilation, so more CO_2 is produced and transported through the internal system of interconnected gas lacunae in plants. The positive correlation between soil redox reactions and CO_2 emission is consistent with this result.

CO_2 emission varied seasonally, increasing with rice growth and temperature. Temperature controls CO_2 production and emission by not only increasing soil microbial activity (Vogel et al., 2014) or controlling their physiology (aka carbon use efficiency) (Allison et al., 2010; Frey et al., ; Kivlin et al., 2013; Sihi et al., 2017), but also by altering plant respiration (Slot et al., 2013), substrate availability and quality, species composition, water availability and aerobic/anaerobic conditions (Davidson and Janssens, 2006; Inglett et al., 2012; Sihi, 2015; Sihi et al., 2016a; 2016b). Higher temperatures increase soil CO_2 emissions in subtropical wetlands (Inglett et al., 2012). C quality influences primarily SOM decomposition at low temperatures while at high temperatures nutrient availability controls SOM decomposition in subtropical wetlands (Sihi et al., 2016b). In our study we have obtained consistent results with these previous reports. Soil CO_2 concentration in porewater increased with temperature especially in double fertilized soils (Figs. 3 and 4), showing higher organic matter decomposition with temperature mainly in double N fertilized soils. Given that the major fraction of rice croplands in southeastern Asia corresponds to puddled/wetland conditions, understanding complex interactions in such environments is important for improving our capacity for future projections under warming climate for both natural and agricultural systems.

Effects of fertilization on CH_4 emissions

Mean CH_4 emissions were 20.4% and 39.5% higher for the standard and double fertilizations than the control, respectively. As stated above, fertilization promotes the deposition of photosynthetically derived C into SOC pools (Ge et al., 2015). Such C can contribute up to 52% of the CH_4 emissions from paddy soils by the exudation of labile organic C from roots to the rhizosphere, which will then produce methane. These results are consistent with the lack of fertilization effects on rice yield. While fertilization can enhance photosynthesis, more photosynthates are allocated to root exudates and this reduces allocation to growth and yield. The other 48% of the CH_4 is emitted from old soil C (Minoda et al., 1996), promoting CH_4 production and emissions (Minoda et al., 1996). Fertilization, especially N fertilization, will also increase the availability of nutrients, which will promote CH_4 production and emissions from microbes (Naik et al., 2015). Since N fertilizer was provided in NH_4^+ form, it could have inhibited CH_4 oxidation because of the structural resemblance of NH_4^+ with CH_4 promoting enzymatic substrate competition (Gulledge and Schimel, 1998). Nevertheless, most studies testing for different methods and substances for N fertilization have observed enhancement of CH_4 production and emission (Pathak, 2010), which is in agreement with our results.

CH_4 emission varied seasonally and emissions were low soon after rice transplantation when soil was not strictly anaerobic. The correlation of soil redox reactions with CH_4 emission supported this finding (Table S3). CH_4 emissions were also low during the final ripening and drainage periods. These results agreed with those by Minamikawa et al. (2014), in which a lowering of the water table decreased the abundance of the methanogenic archaeal population and hence CH_4 production and increased the abundance of methanotrophs and thus CH_4 oxidation.

Effects of fertilization on N_2O emissions

N_2O emission was low throughout the growing season, with no obvious pattern of seasonal variation (Fig. X). The paddies in our study region are strongly N limited (Wang et al., 2015), so together with low levels of soil O_2, most of the N_2O produced is likely reduced to N_2 and this would lead to very low emissions or even a net uptake of N_2O (Zhang et al., 2010). Pulses in ammonium and nitrate availability after a fertilization have been related to N_2O production (Pathak et al., 2002). Specific N_2O fluxes and the contribution of nitrifying and denitrifying bacteria are controlled mainly by soil moisture (Davidson et al., 1993). However, the results of our study showed that the N added by fertilization has not been sufficient to rise N_2O emissions in this paddies with low N concentrations.

Best management practices to reduce GWP

Our results suggested that the application of fertilizer increased the impacts of rice agriculture on climate change, with higher total GWPs per unit yield compared to the controls. The fertilizations did not significantly increase the rice yield but they increased the soil porewater nutrient concentrations, which has the potential risk of nutrient loss, eutrophication and higher costs. Judicious use of fertilization should be reconsidered in a sustainable agriculture and our results provide strong evidence that the current strategy of fertilization in most rice croplands in subtropical China over several years will saturate the soil fertility, increasing the release of nutrients to continental water and favoring CH_4 and CO_2 production and emission without increasing rice yield. Our findings suggest that alternating years of standard and low fertilizations could decrease water pollution and mitigate GHG emissions without decreasing rice yield, an issue to be further studied.

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Tables

Table 1. Results of the linear analysis of the effects of the mixed models, with treatment as a fixed factor, plot and time as random factors on greenhouse-gas emissions and porewater greenhouse-gas concentrations and other soil variables as dependent variables. R^2_m is the variance explained by the fixed factors, and R^2_c is the variance explained by the overall model (fixed + random). Statistical significant values are in bold type.

Dependent variable	Y~ treatment, random=list(~1|plot, ~1|time)				
	R^2	Standard fertilization (relative to control)		Double fertilization (relative to control)	
		t	*P*	*t*	*P*
CO_2 emission	**R^2_m=0.0036, R^2_c=0.92**	**41.8**	**<0.0001**	**2.51**	**0.088**
CH_4 emission	R^2_m=0.024, R^2_c=0.44	1.15	0.25	**2.23**	**0.028**
N_2O emission	R^2_m=0.0035, R^2_c=0.49	-0.68	0.5	-0.83	0.41
Dissolved porewater CO_2 concentration	R^2_m=0.21, R^2_c=0.54	-0.71	0.48	**5.93**	**<0.0001**
Dissolved porewater CH_4 concentration	R^2_m=0.0037, R^2_c=0.39	-0.81	0.42	-0.59	0.55
Dissolved porewater N_2O concentration	R^2_m=0.017, R^2_c=0.11	0.43	0.67	1.44	0.15
Soil pH	R^2_m=0.00045, R^2_c=0.99	0.15	0.88	-1.89	0.063
Soil Eh	R^2_m=0.0015, R^2_c=0.97	-2.20	0.031	-1.64	0.10
Soil salinity	R^2_m=0.0328, R^2_c=0.66	1.95	0.055	**3.33**	**0.0013**
Soil temperature	R^2_m=0.00032, R^2_c=0.98	-0.70	0.49	0.73	0.47
Soil water content	R^2_m=0.0012, R^2_c=0.92	1.06	0.29	1.20	0.24
Porewater NH_4^+ concentration	R^2_m=0.043, R^2_c=0.24	1.37	0.17	**2.56**	**0.013**
Porewater NO_3^- concentration	R^2_m=0.0026, R^2_c=0.0026	0.32	0.75	0.55	0.58
Porewater TN concentration	R^2_m=0.018, R^2_c=0.33	0.82	0.41	1.75	0.084
Porewater TP concentration	R^2_m=0.022, R^2_c=0.081	0.34	0.73	1.59	0.12
Porewater DOC concentration	R^2_m=0.0063, R^2_c=0.82	-1.52	0.13	-1.91	0.059

Table 2. Effect of the fertilizations on the global warming potential.

Treatment	Rice yield (Mg ha^{-1})	Global warming potential (kg CO_2-eq ha^{-1})			Global warming potential (kg CO_2-eq ha^{-1})	Global warming potential (kg CO_2-eq Mg^{-1} yield)
		CO_2	CH_4	N_2O		
Control	7.07±0.40	19286±2553	2607±937	-27.4±84.9	21801±3326	3134±616
Standard fertilization	7.44±0.35	21883±1009	3139±340	-44.7±52.6	24872±1147	3363±245
Double fertilization	6.97±0.24	20709±681	3635±722	-55.1±44.5	24160±295	3475±129

Figure legends

Figure 1. Seasonal variation of CO_2 (A) and CH_4 (B) emissions and N_2O (C) fluxes from the control and treatment plots. Error bars indicate one standard error of the mean of triplicate measurements.

Figure 2. Seasonal variation of dissolved porewater CO_2 (A), CH_4 (B) and N_2O (C) concentrations for the control and treatment plots. Error bars indicate one standard error of the mean of triplicate measurements.

Figure 3. Seasonal variation of soil pH (A), Eh (B), temperature (C), salinity (D) and water content (E) for the control and treatment plots. Error bars indicate one standard error of the mean of triplicate measurements.

Figure 4. Seasonal variation of dissolved porewater NH_4^+ (A), NO_3^- (B), TN (C), TP (D) and DOC (E) concentrations for the control and treatment plots. Error bars indicate one standard error of the mean of triplicate measurements.

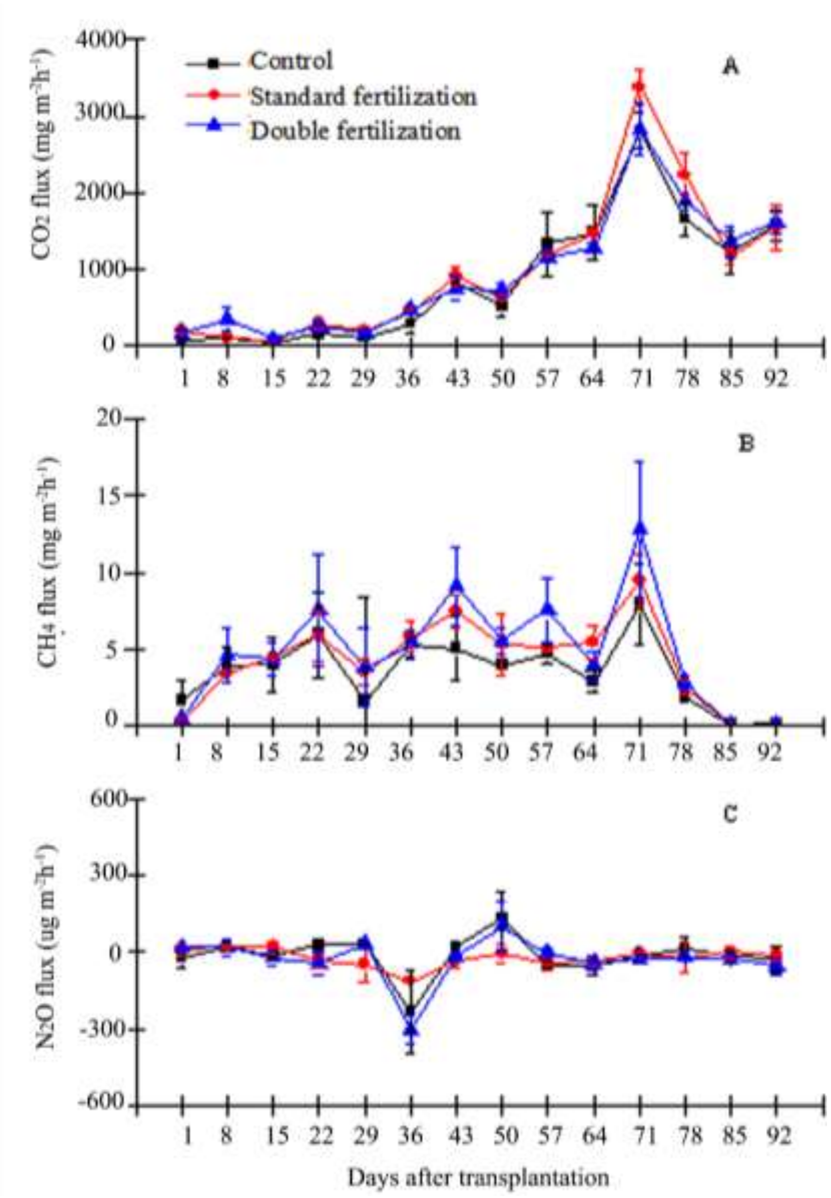
A
B
C
Control
Standard fertilization
Double fertilization
CO2 flux (mg m-2h-1)
4000
3000
2000
1000
0
CH4 flux (mg m-2h-1)
20
15
10
5
0
N2O flux (ug m-2h-1)
600
300
0
-300
-600
1
8
15
22
29
36
43
50
57
64
71
78
85
92
Days after transplantation

Figure 1

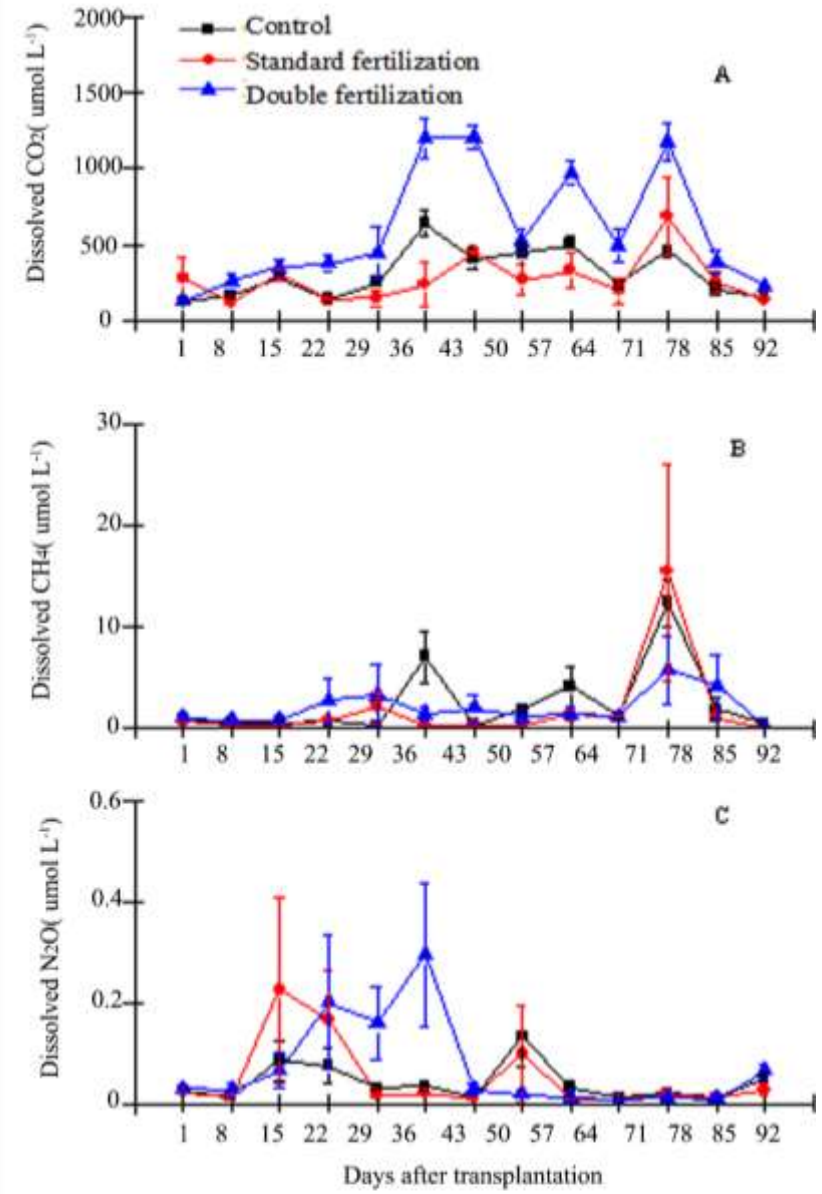
Control
Standard fertilization
Double fertilization
A
Dissolved CO2(umol L-1)
2000
1500
1000
500
0
B
Dissolved CH4(umol L-1)
30
20
10
0
C
Dissolved N2O(umol L-1)
0.6
0.4
0.2
0
1 8 15 22 29 36 43 50 57 64 71 78 85 92
Days after transplantation

Figure 2

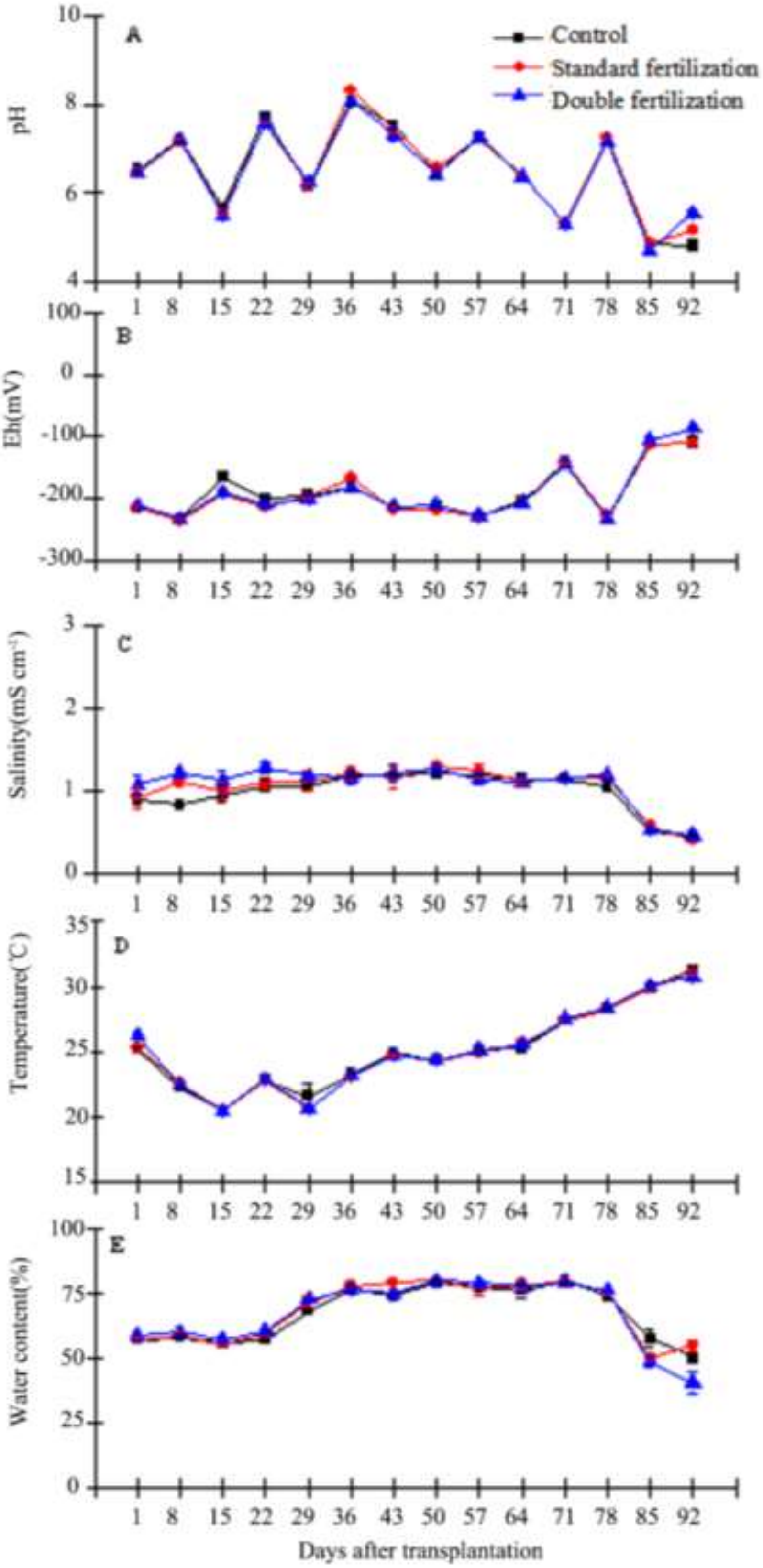
A
B
C
D
E
Control
Standard fertilization
Double fertilization
pH
10
8
6
4
Eh(mV)
100
0
-100
-200
-300
Salinity(mS cm⁻¹)
3
2
1
0
Temperature(°C)
35
30
25
20
15
Water content(%)
100
75
50
25
0
1
8
15
22
29
36
43
50
57
64
71
78
85
92
Days after transplantation

Figure 3

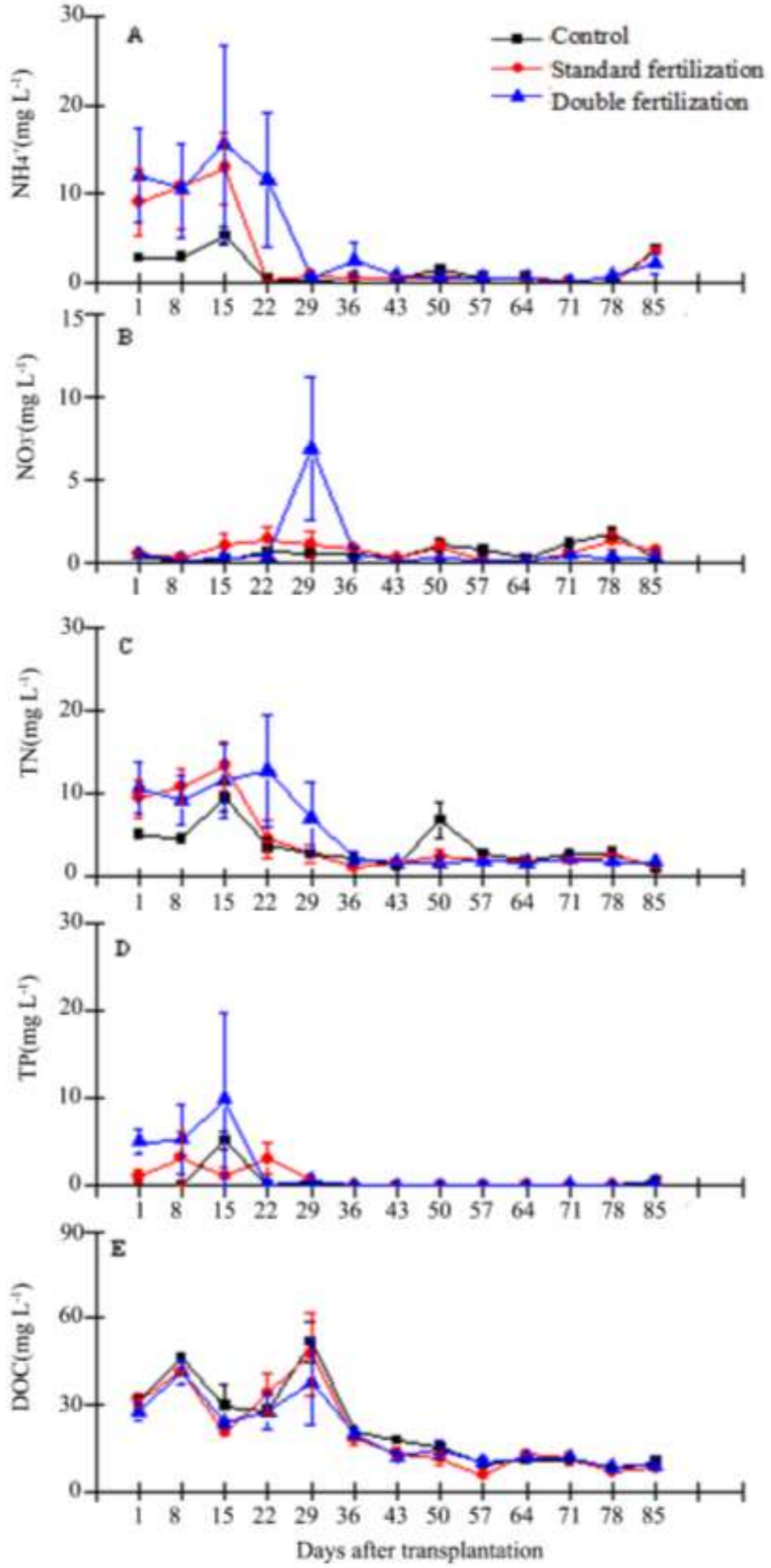A
B
C
D
E
Control
Standard fertilization
Double fertilization
NH4+(mg L-1)
NO3-(mg L-1)
TN(mg L-1)
TP(mg L-1)
DOC(mg L-1)
0
10
20
30
5
15
60
90
1
8
15
22
29
36
43
50
57
64
71
78
85
Days after transplantation

Figure 4