**Dear editor and reviewers:** Thank you very much for your patience and your great support! We have corrected our manuscript according to your valuable comments. Please see the point-to-point answers and tracking manuscript. Thank you very much!

Review of the manuscript BG- 2015-478 (revised version 4) entitled: 'Global warming potential and greenhouse gas intensity in rice agriculture driven by high yields and nitrogen use efficiency: A 5-year field study'

I reviewed the previous version of the manuscript. Compared with that version, this current version (v4) is considerably improved. Most of the comments and suggestions made on the previous version have been followed and revised, and I commend authors for their efforts. In general, the responses to my previous comments are adequate, except on two key issues. These two issues are: (1) Whether to call this as a '5-year study', and (2) The inadequacy of explaining the methodology of estimating GHG emissions from 'farm machinery production' in Table 4.

The reasons for my concern relating to above noted two issues are explained below, but I would like to re-iterate first, my full support for the publication of this study, due to the present global priority of developing cropping systems that are capable of increasing food production while minimizing the environmental impact. Therefore, provided that the two key issues I am explaining here are corrected, I recommend this manuscript for publication in the journal Biogeosciences.

A: Thank you very much for your patience and your great support! We have revised our manuscript according to your valuable comments.

## Issue 1: Whether to call this as a '5-year study'

I carefully considered the response provided by authors to my previous comment on this issue; however, I cannot agree with their response due to following reasons:

In lines 78 to 80 in the manuscript, authors have stated that: 'In this study, we evaluated GWP and GHGI of rice-wheat crop rotation managed under several scenarios of ISSM by taking CO2 equivalent emissions from all sources and sinks into account for 5 years.'

However, what follows in the statistical analysis (Table 3) do not support this statement. Statistical analysis included only three years of data. Presentation of the results in the remaining Tables and Figures was also limited for only three years of data. Furthermore, following text in the manuscript indicates clear contradictions with the above statement:

Line 203: During the three cropping rotations...... rice and wheat yields...

Line 205: On average over the three cycles, the annual rice yield...

Line 212: ...rice and wheat yields from the three years were not...

Line 225: ... During the three annual rice-wheat rotations.....CH4 fluxes ranged from...

Line 229: ... Temporal variation was significant during the three cycles (Table...

Line 263: Across the three years ISSM-N1 and ISSM-N2...

Line 323: During the three years, the annual cumulative CH4 emissions...

Considering these contradictions it is difficult to agree with author's explanation. As the discussion and conclusions of the manuscript are largely (I would say 90%) based on three years of data, it is not reasonable to call it as a '5-year' study.

I suggest authors to consider following revisions in order to minimize these obvious contradictions: (a) the title of the manuscript could be revised as: 'Global warming potential and greenhouse gas intensity in rice agriculture driven by high yields and nitrogen use efficiency'; and (b) Particular places of the text that put emphasis on the phrase: '5-year study' (e.g. reference to '5- year' in Line 17) should be revised.

A: Thank you very much for your comment. We have removed the term "A 5-year field study" from the title and revised the manuscript accordingly.

# Issue 2: Methodology of estimating GHG emissions associated with farm machinery production:

In response to my previous comment on this issue, authors have responded saying: 'We used electricity energy units of kilowatt hour (0.0725 kg CE/kg active ingredient) for calculating CO2 emissions from farm machinery production as presented in Table 1 of Lal 2004.'

This explanation is not adequate. The above emission factor (from Table 1, Lal 2004) is simply the one used for calculating the CO2 emissions per unit of electricity consumed.

Estimating GHG emissions associated with farm machinery production is somewhat complex and involves several steps that need to be explained adequately. For example: Carbon dioxide equivalent (CE) emissions associated with the production of particular piece of farm machinery are influenced by following parameters:

- (a) Carbon equivalent emission factor (EF) for producing a unit weight of machinery (CO2eq/kg machinery),
- (b) Average weight (W) of the piece of farm machinery (kg),
- (c) The fraction of machine life used for a particular farm operation (Fraction).

For calculating the item 'c' above, two parameters are needed: (d) Average life span of the piece of machinery (hours), and (e) Time, this piece of machinery is used for a given field operation (e.g. tillage or planting) (hours/ha)

Authors have not sufficiently explained any of these parameters, steps and any source reference for how different values of 'kg active ingredient/ha' under the column: 'farm machinery production' in Table 4 was obtained. The newly added 'supplementary resources 2' provides only a seasonal breakdown of the data already presented in the upper part of the Table 4 and do not provide any additional information.

I suggest authors to briefly explain, how these values were derived in the supplementary resource 2, since the methodology for this type of emission reporting need to be transparent.

A: We appreciate for your nice patience. You are right that estimating GHG emissions associated with farm machinery production is quite complex and involves several. Actually, we did not intend to calculate the GHG emissions associated with farm machinery production as in our previous Table 4. We actually intend to calculate the GHG emissions from electricity energy consumed associated with farm operation, i.e. threshing in this case study. We used electricity energy coefficients (0.0725 kg CE

per kilowatt hour) for calculating  $CO_2$  emissions from threshing as presented in the revised Table 4. Electricity energy consumption was calculated according to the power and working hours of thresher. The power of thresher is 15 kilowatt in this experiment. Revised accordingly Table 4 and Supplementary resources 2.

In addition to two key issues mentioned above, following minor comments (mostly editorial corrections) need to be corrected.

#### Minor corrections that needs to be corrected:

Line 58: 'into' is one word. Not 'in to'

A: Revised accordingly Page 3, Line 58.

Line 112: ...and N supplied from rapeseed cake in...

A: Revised accordingly Page 5, Line 112.

Line 121: ...rapeseed cake manure was applied for the rice crop.

A: Revised accordingly Page 5, Line 121.

Line 146: ... The CH4 and N2O fluxes were calculated

A: Revised accordingly Page 6, Line 146.

Line 147: ... over time as described by Jia et al. (2012).

A: Revised accordingly Page 6, Line 147.

Line 175: ...N, P, and K fertilizer...

A: Revised accordingly Page 7, Line 175.

Line 178: ... 1.3 kg C equivalent kg-1 N (Lal, 2004).

A: Revised accordingly Page 7, Line 178.

Line 184-185: '...no specific coefficients were available for China.' Or '... no specific coefficients were available for local conditions'.

A: Revised accordingly Page 7, Line 185.

Line 188-189: Chemical fertilizer was hand broadcasted for each fertilization event.

A: Revised accordingly Page 8, Line 189.

Line 190: ...crop seasons are presented

A: Revised accordingly Page 8, Line 190.

Line 194-195: One-way analysis of variance was conducted to compare the cumulative fluxes of CH4 and N2O, and grain...

A: Revised accordingly Page 8, Line 195-196.

Line 252: Should be as: Irrigation was the second largest source o

A: Revised accordingly Page 10, Line 252.

Line 298: ...fertilizer...

A: Revised accordingly Page 11, Line 298.

Line 316: should be:... period could be due to reduced N losses by leaching and volatilization...

A: Revised accordingly Page 12, Line 316.

Line 391: The decrease is 14% and 18% relative to GHGI in FP scenario. Please replace the word: 'dramatically' with the word 'significantly'. Should read as: Compared with the FP, the ISSM-N1 and ISSM-N2 scenarios significantly reduced the GHGI, ...

A: Revised accordingly Page 14, Line 391.

Line 405: Above ground crop residue was removed in this study. Therefore, this sentence should be

revised. For example, you may say as: This may be due to the incorporation of rapeseed cake and enhanced below-ground crop residue associated with higher crop productivity (Ma et al., 2013).

A: Revised accordingly Page 15, Line 405-406.

Line 418: ...in the future, because...

A: Revised accordingly Page 15, Line 418.

Line 423: Should read as: Of the two crops, CH4 and irrigation were important for rice, but less important for wheat, in which N2O losses were expected to...

A: Revised accordingly Page 15, Line 423.

Line 442: Should read as: ...increasing grain yields and at the same time reducing the substantial environmental impact of intensive agriculture...

A: Revised accordingly Page 16, Line 442.

Line 466: ...GHGI was lowered by 23%.

A: Revised accordingly Page 17, Line 466.

### Corrections that should be done for Table 4:

Please indicate, what is the active ingredient for each input category, immediately below the heading as you have already done for irrigation water (cm). For example: Tillage ((kg diesel/ha), planting (kg diesel/ha)....

A: Thank you for your comment. Revised accordingly Table 4 and Supplementary resources 2.

### **Corrections that should be done for Table 5:**

Please check the negative or positive symbols for the value of SOCR in Table 5. I made this comment before; however, it is still not corrected. If the soil is a sink for C, the value should be negative. If the soil is a source of C, the value should to be positive. (SUM of values of each GHG category for each scenario would not yield the value of GWP you have presented in Table 5 at present, because the negative/positive symbols you have put for SOCSR at present is not correct).

A: Thank you for your comment. GWP =  $28 \times CH_4 + 265 \times N_2O + Ei + Eo - 44/12 \times SOCSR$ . We use a minus for SOCSR in the equation to emphasize the soil carbon sequestration effect. Thus, we provide different meaning for SOCSR from your understanding. The SOCSR is calculated as equation: SOCSR =  $(SOC_t - SOC_0) / T \times \gamma \times (1 - \delta 2mm/100) \times 20 \times 10^{-1}$ . Thus, negative value  $(SOC_t < SOC_0)$  indicates soil as a source (not sink) for  $CO_2$  emissions from SOC loss and a positive value  $(SOC_t > SOC_0)$  indicates soil as a sink for C sequestration (not source) as presented in Table 5. Thank you for your patience.

Thank you very much once again for all of your nice comments and great support! Sincerely yours,

Zhengqin (on behalf of all authors)

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Global warming potential and greenhouse gas intensity in rice agriculture driven by 1 high yields and nitrogen use efficiency: A 5-year field study 2 3 4 Xiaoxu Zhang<sup>a</sup>, Xin Xu<sup>a</sup>, Yinglie Liu<sup>a</sup>, Jinyang Wang<sup>a,b</sup>, Zhengqin Xiong<sup>a,\*</sup> 5 <sup>a</sup> Jiangsu Key Laboratory of Low Carbon Agriculture and GHGs Mitigation, College of 6 Resources and Environmental Sciences, Nanjing Agricultural University, Nanjing, 210095, 7 8 China <sup>b</sup> State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese 9 Academy of Sciences, Nanjing 210008, China 10 11 \*Corresponding author (Z. Xiong): 12 13 Tel: +86-25-84395148, Fax: +86-25-84395210, 14 15 E-mail: zqxiong@njau.edu.cn

Abstract: Our understanding of how global warming potential (GWP) and greenhouse gas intensity (GHGI) is affected by management practices aimed at food security with respect to rice agriculture remains limited. In the present study, a 5 year field experiment was conducted in China to evaluate the effects of integrated soil-crop system management (ISSM) mainly consisting of different nitrogen (N) fertilization rates and split, manure, Zn and Na<sub>2</sub>SiO<sub>3</sub> fertilization and planting density on GWP and GHGI after accounting for carbon dioxide (CO2) equivalent emissions from all sources including methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) emissions, agrochemical inputs and farm operations and sinks (i.e., soil organic carbon sequestration). For the improvement of rice yield and agronomic nitrogen use efficiency (NUE), four ISSM scenarios consisting of different chemical N rates relative to the local farmers' practice (FP) rate were carried out, namely, ISSM-N1 (25% reduction), ISSM-N2 (10% reduction), ISSM-N3 (FP rate) and ISSM-N4 (25% increase). The results showed that compared with the FP, the four ISSM scenarios significantly increased the rice yields by 10, 16, 28 and 41% and the agronomic NUE by 75, 67, 35 and 40%, respectively. In addition, compared with the FP, the ISSM-N1 and ISSM-N2 scenarios significantly reduced the GHGI by 14 and 18%, respectively, despite similar GWPs. The ISSM-N3 and ISSM-N4 scenarios remarkably increased the GWP and GHGI by an average of 69 and 39%, respectively. In conclusion, the ISSM strategies are promising for both food security and environmental protection, and the ISSM scenario of ISSM-N2 is the optimal strategy to realize high yields and high NUE together with low environmental impacts for this agricultural rice field.

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#### 1 Introduction

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Rapid population growth and economic development place a growing pressure on increasing food production (Barrett, 2010). An increase in global crop production of 100% would be necessary to sustain the projected demand for human food and livestock feed in 2050 (Tilman et al., 2011). Rice is the staple food for nearly 50% of the world's people, mainly in Asia (Frolking et al., 2002). According to FAO (2010), approximately 600 million people in Asia-Pacific region are suffering from hunger and malnutrition. With the region's population projected to increase by another billion by mid-century, new approaches to increase food production are needed (Chen et al., 2014). With a limited agricultural land area, the intensive agricultural regions of China are facing serious environmental problems due to large inputs of chemical fertilizer and low nitrogen use efficiency (NUE) (Ju et al., 2009; Makino, 2011). Thus, integrated soil-crop system management (ISSM), which redesigns the whole production system based on the local environment and draws on appropriate fertilizer compounds and application ratios, crop densities and advanced water management regimes, has been advocated and developed to simultaneously increase crop productivity and NUE with low carbon dioxide (CO<sub>2</sub>) equivalent emissions per unit product in China (Chen et al., 2014). The key points of the ISSM are to integrate soil and nutrient management with high-yielding cultivation systems, to integrate the utilization of various nutrient sources and match nutrient supply to crop requirements, and to take all soil quality improvement measures into consideration (Zhang et al., 2011).

Carbon dioxide, methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) are the most important greenhouse gases (GHGs) that contribute to global warming (IPCC, 2013). The concept of global warming potential (GWP) has been applied to agricultural lands by taking in–to account of the radiative properties of all GHG emissions associated with agricultural production and soil organic carbon (SOC) sequestration, expressed as CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup> (Robertson and Grace, 2004; Mosier et al., 2006). Although agriculture releases significant amounts of CH<sub>4</sub> and N<sub>2</sub>O into the atmosphere, the net emission of CO<sub>2</sub> equivalents from farming activities can be partly offset by changing agricultural management to increase the soil organic matter content and/or decrease the emissions of CH<sub>4</sub> and N<sub>2</sub>O (Mosier et al., 2006; Smith et al., 2008). If global agricultural techniques are improved, the mitigation potential of

agriculture (excluding fossil fuel offsets from biomass) is estimated to be approximately 5.5–6.0 Pg CO<sub>2</sub> eq. yr<sup>-1</sup> by 2030 (Smith et al., 2008). However, the release of CO<sub>2</sub> during the manufacturing and application of N fertilizer to crops and from fuel used in machines for farm operations can counteract these mitigation efforts (West and Marland, 2002). Therefore, when determining the GWP of agroecosystems, there is a need to account for all sources of GHG emissions, including the emissions associated with agrochemical inputs (Ei) and farm operations (Eo) and sinks, e.g. soil organic carbon (SOC) sequestration (Sainju et al., 2014).

Information on the effects of ISSM scenarios on GWP and greenhouse gas intensity (GHGI) of agricultural systems is limited in China (Ma et al., 2013; Liu et al., 2015). The annual rotation of summer rice-upland crop is a dominant cropping system in China. Previous studies were mainly focused on the initial influences of ISSM practices on CH<sub>4</sub> and N<sub>2</sub>O emissions, but did not account for the contributions of CO<sub>2</sub> emissions from Ei and Eo (Ma et al., 2013; Zhang et al., 2014). In this study, we evaluated GWP and GHGI of rice-wheat crop rotation managed under several scenarios of ISSM by taking CO<sub>2</sub> equivalents emissions from all sources and sinks into account for 5–3 years. We hypothesized that the ISSM strategies would reduce the overall GWP and GHGI compared with local farmers' practices (FP). The specific objectives of this study were to (i) evaluate the effects of different ISSM scenarios on GWP and GHGI; (ii) determine the main sources of GWP and GHGI in a rice-wheat cropping system; and (iii) elucidate the overall performance for each ISSM scenario for different targets to increase grain yields and NUE and reduce GWP and GHGI.

#### 2 Materials and Methods

2.1 Experimental site

A 5-year-field experiment was conducted at the Changshu agro-ecological experimental station (31°32′93″N, 120°41′88″E) in Jiangsu Province, China. This is a typical, intensively managed agricultural area where the cropping regime is dominated by a flooded rice (*Oryza sativa* L.)-drained wheat (*Triticum aestivum* L.) rotation system. The site is characterized by a subtropical humid monsoon climate with a mean annual air temperature of 15.6, 15.2 and 15.8 °C and precipitation of 878, 1163 and 984 mm for three years, respectively. The soil of the field is classified as an *Anthrosol* with a sandy loam texture of 6% sand (1–0.05 mm), 80% silt (0.05–0.001 mm), and 14% clay (<0.001 mm), which developed from lacustrine sediment.

The major properties of the soil at 0–20 cm can be described as follows: bulk density, 1.11 g cm<sup>-3</sup>; pH, 7.35; organic matter content, 35.0 g kg<sup>-1</sup>; and total N, 2.1 g kg<sup>-1</sup>. The daily mean air temperatures and precipitation during the study period from June 15, 2011, to June 15, 2014, are given in the supplementary resource 1.

2.2 Experimental design and management

A completely randomized block design was established in 2009 with four replicates of six treatments, including no nitrogen (NN) and FP as controls, and four ISSM scenarios at different chemical N fertilizer application rates relative to the local FP rate (300 kg N ha<sup>-1</sup>), namely ISSM-N1 (25% reduction), ISSM-N2 (10% reduction), ISSM-N3 (FP rate) and ISSM-N4 (25% increase). The designed ISSM scenarios (only for rice but not for wheat) included a redesigned split N fertilizer application, a balanced fertilizer application that included sodium silicate, zinc sulphate, rapeseed cake (C/N=8) providing an additional 112.5 kg N ha<sup>-1</sup>, and additional phosphorus and potassium, and different transplanting densities, used as the main techniques for improving rice yield and agronomic NUE. The agronomic NUE was calculated as the difference in grain yield between the plots that received N application and the NN plot, divided by the total N rate which included chemical N fertilizer and N supplied from rapeseed cake in the ISSM-N3 and ISSM-N4 scenarios. The details of the fertilizer applications, irrigation, and field management practices of the six different treatments are presented in Table 1. Further information was described previously (Zhang et al., 2014). Each plot was 6 m × 7 m in size with an independent drainage/irrigation system.

One midseason drainage (about one week) and final drainage before harvest were used during the rice-growing season, whereas the plots only received precipitation during the wheat-growing season. The N fertilizer was split into a 6:2:0:2 or 5:1:2:2 ratio of basal fertilizer and topdressings for the rice crop and a 6:1:3 ratio for the wheat crop. All of phosphorous (P), silicon (Si), zinc (Zn) were applied as basal fertilizers for both crops and rapeseed cake manure was applied for the rice crop. Potassium (K) was added as a split (1:1) application to the rice crop and all as basal fertilizer for the wheat crop. The basal fertilization occurred at the time of rice transplanting and wheat seeding. The topdressing was applied at the tillering, elongation and panicle stages of the rice crop and at the seedling establishment and elongation stages of the wheat crop. Aboveground biomass including crop grains and

straws were removed out of the fields for all the treatments.

2.3 Gas sampling and measurements

We measured the  $CH_4$  and  $N_2O$  emissions from each plot of the field experiment over five annual cycles from the 2009 rice-growing season to the 2014 wheat-growing season. The initial 2-yr measurements during the 2009–2011 rice-wheat rotational systems were described in our previous study (Ma et al., 2013). Emissions were measured manually using the static-opaque chamber method. Each replicate plot was equipped with a chamber with a size of 50 cm  $\times$  50 cm  $\times$  50 cm or 50 cm  $\times$  50 cm  $\times$  110 cm, depending on the crop growth and plant height. The chamber was placed on a fixed PVC frame in each plot and wrapped with a layer of sponge and aluminum foil to minimize air temperature changes inside the chamber during the period of sampling. Gas samples were collected from 9:00 to 11:00 am using an airtight syringe with a 20-ml volume at intervals of 10 min (0, 10, 20 and 30 min after chamber closure). The fluxes were measured once a week and more frequently after fertilizer application or a change in soil moisture.

The gas samples were analyzed for  $CH_4$  and  $N_2O$  concentrations using a gas chromatograph (Agilent 7890A, Shanghai, China) equipped with two detectors. Methane was detected using a hydrogen flame ionization detector (FID), and  $N_2O$  was detected using an electron capture detector (ECD). Argon-methane (5%) and  $N_2$  were used as the carrier gas at a flow rate of 40 ml min<sup>-1</sup> for  $N_2O$  and  $CH_4$  analysis, respectively. The temperatures for the column and ECD detector were maintained at 40 °C and 300 °C, respectively. The oven and FID were operated at 50 °C and 300 °C, respectively. The  $CH_4$  and  $N_2O$  fluxes—rate were calculated using a linear increase in the two gas concentrations over time <u>as</u> described by Jia et al. (2012).

150 2.4 Topsoil organic carbon sequestration measurements

To measure the organic carbon content of the topsoil as described by Zhang et al. (2014), soil samples were collected after the wheat harvest in 2009 and 2014 from all experimental plots at a plowing depth of 0–20 cm. The soil organic carbon sequestration rates (SOCSR) were calculated as follows (Liu et al., 2015):

SOCSR (t C ha<sup>-1</sup> yr<sup>-1</sup>) = (SOC<sub>t</sub> – SOC<sub>0</sub>) / T × 
$$\gamma$$
 × (1 –  $\delta_{2mm}$ /100) × 20 × 10<sup>-1</sup> (1)

In Eq. (1), SOC<sub>t</sub> (g C kg<sup>-1</sup>) and SOC<sub>0</sub> (g C kg<sup>-1</sup>) are the SOC contents measured in the

soils sampled after the wheat was harvested in 2014 and 2009, respectively. T refers to the experimental period (yr).  $\gamma$  and  $\delta_{2mm}$  are the average bulk density and the gravel content (>2 mm) of the topsoil (0–20 cm), respectively.

### 2.5 GWP and GHGI measurements

To better understand the overall GHG impact of the rice-wheat crop rotation managed under different ISSM scenarios, the GWP and GHGI were calculated as the following equations (Myhre et al., 2013):

164 GWP (kg CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup>) =  $28 \times CH_4 + 265 \times N_2O + Ei + Eo - 44/12 \times SOCSR$  (2)

GHGI (kg  $CO_2$  eq. kg<sup>-1</sup> grain yield yr<sup>-1</sup>) = GWP/grain yield (3)

In Eq. (2), Ei (kg CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup>), Eo (kg CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup>) and SOCSR (kg C ha<sup>-1</sup> yr<sup>-1</sup>) represent CO<sub>2</sub> equivalent emissions from the agrochemical inputs, farm operations and soil organic carbon sequestration rate, respectively. The global warming potential of 1 kg CH<sub>4</sub> and 1 kg N<sub>2</sub>O are 28 and 265 kg CO<sub>2</sub> equivalents respectively (without inclusion of climate-carbon feedbacks), based on 100-yr time scale (Myhre et al., 2013). 12 and 44 refers to molecular weights of C and CO<sub>2</sub>, respectively. The grain yield is expressed as the air-dried grain yield.

Therefore, the GWP of the cropland ecosystem equals the total CO<sub>2</sub> equivalent emissions minus the SOC change per unit land area. In addition to CH<sub>4</sub> and N<sub>2</sub>O emissions, we considered CO<sub>2</sub> equivalent emissions associated with the use of agrochemical inputs (Ei), such as the manufacture and transportation of the N, P<sub>2</sub> and K fertilizers (Snyder et al., 2009), and farm operations (Eo), such as the water used for irrigation (Zhang et al., 2013) and diesel fuel (Huang et al., 2013a). The CO<sub>2</sub> equivalent emissions of N fertilizer were calculated as the mean value of the C emissions of 1.3 kg C equivalent kg<sup>-1</sup> N (Lal, 2004). Similarly, the CO<sub>2</sub> equivalent for irrigation was calculated from the total amount of water used during the rice-growing season; the coefficient for the C cost was 5.16 (kg C eq. cm<sup>-1</sup> ha<sup>-1</sup>) originated from the value of 257.8 kg C eq. ha<sup>-1</sup> for a 50 cm of irrigation provided by Lal (2004). The CO<sub>2</sub> equivalents of other Ei (P and K fertilization, manure, herbicide, pesticide and fungicide applications) and Eo (tillage, planting, harvest, and threshingfarm machinery production) were recorded and also estimated by coefficients provided by Lal (2004) since no specific coefficients were available for local conditions. We collected the data specific to China's

187 fertilizer manufacture and consumption, and obtained the C emission coefficients to be 0.07 and 0.1 kg C eq. kg<sup>-1</sup> of active ingredient for Si and Zn fertilizer, respectively. The C emission 188 factor for these farm operations depends on diesel used as fuel or electricity. Chemical 189 190 fertilizers wasere hand spraying broadcasted for each fertilization event. Detailed information 191 of each Ei and Eo component for rice and wheat crop seasons was are presented in 192 Supplementary resource 2. 193 2.6 Statistical analysis Repeated-measures multivariate analysis of variance (MANOVA) and linear relationships 194 were determined using JMP 7.0, ver. 7.0 (SAS Institute, USA, 2007). The F-test was applied 195 to determine whether there were significant differences among practices, years and their 196 197 interaction at P < 0.05. One-way analysis of variance was conducted to compare the cumulative fluxes determine the emissions of CH<sub>4</sub> and N<sub>2</sub>O, and the grain yield among the 198 199 different treatments. Tukey's HSD test was used to determine whether significant differences 200 occurred between the treatments at a level of P < 0.05. Normal distribution and variance 201 uniformity were checked and all data were consistent with the variance uniformity (P > 0.05)202 within each group. The results are presented as the means and standard deviation (mean  $\pm$  SD, 203 n = 4).

## 3 Results

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205 3.1 Crop production and agronomic NUE

During the three cropping rotations from 2011 to 2014, the rice and wheat yields varied significantly among the treatments (Table 2). The grain yields ranged from 5.83 to 12.11 t ha<sup>-1</sup> for rice and 1.75 to 6.14 t ha<sup>-1</sup> for wheat. On average over the three cycles, the annual rice yield of the FP was significantly lower than that of the ISSM scenarios of ISSM-N1, ISSM-N2, ISSM-N3 and ISSM-N4. Compared with the FP, rice grain yields increased by 10% and 16% for the ISSM-N1 and ISSM-N2 scenarios, respectively, i.e., with the lower N input, by 28% for the ISSM-N3 scenario with the same N input and by 41% for the ISSM-N4 scenario with the highest N input. However, we did not observe any significant increases in the wheat-grain yields compared with the FP except for the ISSM-N4 scenario. Statistical analysis indicated that rice and wheat yields from the three years were not significantly influenced by the interaction of cultivation patterns and cropping year (Table 3).

217 The agronomic NUE for the rice and wheat of the fertilized plots ranged from 9.2 to 16.1 and 19.5 to 24.7 kg grain kg  $N^{-1}$ , respectively (Fig. 1). The higher NUE in the wheat season 218 219 was mainly due to the relatively lower N fertilizer (40%) rates used for wheat compared with 220 that for rice. As expected, the rice agronomic NUE significantly increased by 75, 67, 35 and 221 40% for the ISSM-N1, ISSM-N2, ISSM-N3 and ISSM-N4 scenarios, respectively, compared 222 with the FP (Fig. 1). For the wheat crop, the agronomic NUE increased by 12 and 14% in the 223 ISSM-N1 and ISSM-N2 scenarios, respectively, and slightly decreased in the ISSM-N3 and 224 ISSM-N4 scenarios compared with the FP, mainly because the current ISSM strategy was 225 only designed for rice and not wheat production. 226 3.2 CH<sub>4</sub> and N<sub>2</sub>O emissions 227 All plots showed similar CH<sub>4</sub> emission patterns, being a source in the rice season and 228 negligible in the wheat season (Fig. 2). During the three annual rice-wheat rotations from 2011 to 2014, the CH<sub>4</sub> fluxes ranged from -3.89 to 99.67 mg C m<sup>-2</sup> h<sup>-1</sup>. The seasonal CH<sub>4</sub> 229 230 emissions varied significantly among the treatments during the rice-growing season (Table 3, 231 Fig. 2). No significant difference was found between the FP, ISSM-N1 and ISSM-N2 plots. Temporal variation was significant during the three cycles (Table 3, P < 0.001). Averaged 232 233 across years, the CH<sub>4</sub> emission was greater in the ISSM-N3 and ISSM-N4 plots than in the NN, FP, ISSM-N1 and ISSM-N2 plots (Table 2, P < 0.05). However, compared with the NN 234 plots, the FP, ISSM-N1 and ISSM-N2 plots with inorganic fertilizer application resulted in 235 increased CH<sub>4</sub> emission rates of 59.9, 41.9 and 43.0%, respectively, averaged over the 236 237 rice-growing seasons. The CH<sub>4</sub> emission rates were further enhanced by 198.5% in the ISSM-N3 plots and by 246.7% in the ISSM-N4 plots. 238 The annual N<sub>2</sub>O fluxes varied from -33.1 to 647.5 µg N<sub>2</sub>O-N m<sup>-2</sup> h<sup>-1</sup>, with most N<sub>2</sub>O 239 240 emissions occurring during the wheat-growing season after fertilization events, and several small emission peaks during the rice-growing season (Fig. 3). With respect to the N 241 242 application effect, the annual cumulative N<sub>2</sub>O emissions for all four ISSM scenarios were significantly higher than that in NN (P < 0.05). Relative to the FP plot, the ISSM-N1 and 243 244 ISSM-N2 scenarios decreased the annual N<sub>2</sub>O emissions by an average of 41% and 22%, respectively (Table 2). The ISSM-N4 scenario significantly increased the cumulative N<sub>2</sub>O 245

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247 higher than that in FP) and additional N via manure application compared to the FP practice, 248 although there was no significant difference between the ISSM-N3 and FP plots. 3.3 Annual GWP and GHGI 249 250 Based on the perspective of the carbon footprint, we included the GHG emissions associated 251 with all of the inputs (Ei and Eo), and SOC sequestration was expressed as kg CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup>. The CO<sub>2</sub> equivalent emissions associated with Ei and Eo are presented in Table 4. The 252 253 CO<sub>2</sub> equivalents rates from N fertilizer dominated not only the chemical input section (67–75% 254 of Ei) but also the total CO<sub>2</sub> equivalents from agricultural management (45–50% of the sum 255 of the Ei and Eo). And I rrigation was the second largest source of CO2 equivalents associated with agricultural management after N fertilizer (19-31% of the sum of the Ei and Eo). The 256 GWP ranged from 8425 to 22711 kg CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup> for the NN and the ISSM-N4 plots, 257 respectively (Table 5). Although fertilized treatments increased the annual CH<sub>4</sub> and N<sub>2</sub>O 258 259 emissions in comparison with the NN plot, it also increased the SOC sequestration in these 260 cropping systems. Of the main field GHGs that were directly emitted, CH<sub>4</sub> accounted for 59-78% of the GWP in all plots. An increase in the annual SOC content led to a significant 261 decrease in the GWP (contributed 5-9% decrease of the GWP except in the NN plot). The 262 263 CO<sub>2</sub> equivalents from agricultural management practices, emissions associated with Ei (2493–4300 CO<sub>2</sub> eq. ha<sup>-1</sup> yr<sup>-1</sup>) were higher than those associated with Eo (1296–1708 CO<sub>2</sub> eq. 264 ha<sup>-1</sup> yr<sup>-1</sup>) in the fertilized plots. There was no significant difference in the annual GWP 265 observed between the FP, ISSM-N1 and ISSM-N2 plots (Table 5). Across the three years, 266 267 ISSM-N1 and ISSM-N2 slightly reduced the GWP by 12 and 10%, respectively; however, ISSM-N3 and ISSM-N4 significantly increased the GWP by an average of 55 and 84%, 268 269 respectively, in comparison with the FP. 270 The GHGI was used to express the relationship between GWP and grain yield. The GHGIs (kg CO<sub>2</sub> eq. t<sup>-1</sup> grain) in this study ranged from 712 to 1245 kg CO<sub>2</sub> eq. t<sup>-1</sup> grain 271 (Table 5). The significant difference in the GHGI of grain was found between the FP and the 272 ISSM strategies. Compared with the FP, ISSM-N1 and ISSM-N2 significantly reduced the 273 274 GHGI by 14 and 18%, respectively, mainly due to the increased grain yield and SOC sequestration as well as reduced GHG emissions for the ISSM strategies of reasonable N 275 fertilizer management and suitable planting density. Although N fertilizer or 276

organic/inorganic combination fertilizer application reduced the SOC losses caused by crop cultivation and increased the grain yields, the GHGIs were generally higher for the ISSM-N3 and ISSM-N4 scenarios than the ISSM-N1 and ISSM-N2 scenarios due to further increases in CH<sub>4</sub> and N<sub>2</sub>O emissions.

#### C114 and N2O emissions

### 4 Discussion

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4.1 Grain yield and agronomic NUE as affected by ISSM strategies

Grain yields are directly related to fertilizer management. The MANOVA results indicated that the rice and wheat grain yields were significantly affected by the cultivation strategies (Table 3, P < 0.001), which is in agreement with previous results (Chen et al., 2011; Zhang et al., 2011). Compared with the FP, rice yields increased significantly by all four ISSM scenarios (Table 2). However, the wheat grain yield decreased significantly when the N fertilizer rate was reduced by 25% (N1 scenario). It has been reported in previous studies that ISSM strategies can effectively improve the rice grain yield (Ma et al., 2013; Liu et al., 2015). First, the adjusted transplanting density for the ISSM-N1, ISSM-N2, and ISSM-N3 scenarios would produce a positive effect on rice yield by influencing rice colony structure, which agreed with Wu et al. (2005). Second, split application of N fertilizer to match crop demand in the ISSM-N1, ISSM-N2, ISSM-N3 and ISSM-N4 scenarios would significantly increase agronomic NUE and rice yield which had been reported previously by Liu et al. (2009). In the present study, ISSM-N1 and ISSM-N2 significantly increased annual rice production by 10 and 16%, respectively, in comparison with the FP (Table 2). This finding is consistent with the results of Peng et al. (2006), who reported that a 30% reduction in the total N rate during the early vegetative stage did not reduce the yield but slightly increased it when combined with the modified farmers' fertilizer practice. Third, integrated management of three macronutrients: N, P and K as well as the two micronutrients: Si and Zn were considered as essential for sustainable high crop yields. Additional Si and Zn fertilizers for the ISSM-N3 and ISSM-N4 scenarios would support better seedling establishment and reduce both biotic and abiotic stress, thus produced higher yields (Wang et al., 2005; Slaton et al., 2005; Kabata-Pendias and Mukherjee, 2007; Hossain et al., 2008). As expected, when the total N rate was at the FP rate and/or increased by 25%, in combination with other ISSM strategies (e.g. rapeseed cake manure, additional P and K, applying Si and Zn fertilizer), the rice yield in these ISSM-N3 and ISSM-N4 plots increased substantially by 28 and 41%, respectively. Based on a long-term fertilizer experiment, Shang et al. (2011) reported that organic fertilizer incorporation significantly increased the early rice grain yield. This may have resulted from the organic fertilizer applied in combination with adequate nutrients contributing to alleviate potential yield limiting factors of rice.

It has been suggested that N losses vary depending on the timing, rate, and method of N application, as well as the source of N fertilizer (Zhu, 1997). In addition to high rates of N and improper timing of N application, rapid N losses (via ammonia volatilization, denitrification, surface runoff, and leaching) are important factors that cause low agronomic NUE of irrigated rice in China (Peng et al., 2006). Compared with the FP plot, the rice agronomic NUE was significantly increased by 75, 67, 35 and 40% under the ISSM-N1, ISSM-N2, ISSM-N3 and ISSM-N4 scenarios, respectively (Fig. 1). The higher rice agronomic NUE in our study over the experimental period could be due to the greatly reduced N losses by leaching and volatilization as well as the improvement of N bioavailability in the rice crop season (Zhao et al., 2015). Organic/inorganic combination fertilizer application also increases uptake by crops compared with the traditional farmers' practice (Peng et al., 2006). These findings suggest that the ISSM strategy is an effective method for improving grain yield and agronomic NUE for future sustainable rice agriculture in China.

4.2 CH<sub>4</sub> and N<sub>2</sub>O emissions as affected by ISSM strategies

During the three years, the annual cumulative  $CH_4$  emissions, on average, varied from 133 to 469 kg C ha<sup>-1</sup>yr<sup>-1</sup> (Table 2), and these values fell within the range of 4.1 to 1015.6 kg  $CH_4$  ha<sup>-1</sup> observed previously in a rice field (Huang et al., 2004). Methane emissions were highest during rice season, but only during the flooding period. Mainly because  $CH_4$  was produced in the anaerobic zones of submerged soils by methanogens and is oxidized into  $CO_2$  by methanotrophs in the aerobic zones of wetland soils and in upland soils (Le Mer and Roger, 2001). The MANOVA results indicated that obvious effects of cultivation paterns and years on  $CH_4$  emissions were found during the rice-wheat rotations (Table 3, P < 0.001). The  $CH_4$  emissions were not significantly affected by the cycles but affected by crop season (Table 5, Fig. 2). In this study, no significant difference in  $CH_4$  emission was observed

between the FP, ISSM-N1 and ISSM-N2 plots. However, compared with the FP plot, the ISSM-N3 and ISSM-N4 scenarios emitted 87 and 118% more CH<sub>4</sub>, respectively (Table 5), which is probably due to the incorporation of the organic rapeseed cake manure. Previous reports support the observations that CH<sub>4</sub> emissions were significantly increased with the application of organic amendments (Ma et al., 2009; Thangarajan et al., 2013; Zou et al., 2005). Apparently, additional application of Si and Zn fertilizers had no significant effect on CH<sub>4</sub> and N<sub>2</sub>O fluxes, which was consistent with the result of Xie et al. (2015). Moreover, rice growth was found to be significantly increased under the ISSM-N3 and ISSM-N4 scenarios. In this case, the organic matter inputs such as root litter and rhizodeposits in the ISSM-N3 and ISSM-N4 scenarios were probably also higher than in the other plots, and thus soil C input, which served as an additional source of substrates for the methanogens in the rice paddies, likely contributed to the increase in CH<sub>4</sub> emissions (Ma et al., 2009). Finally, because the rice plants acted as the main pathway for CH<sub>4</sub> transports from the soil to the atmosphere, the higher biomass may have facilitated more CH<sub>4</sub> emissions (Yan et al., 2005).

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Denitrification and nitrification are the main processes that produce N<sub>2</sub>O in the soil (Paul et al., 1993). The N<sub>2</sub>O emission patterns varied during the rice and wheat growing seasons which were partially associated with the anaerobic conditions prevailing in a rice paddy. Changes in the soil water content strongly influenced the soil N<sub>2</sub>O emissions and resulted in negligible N<sub>2</sub>O emissions when the rice field was flooded (Fig. 3), which is consistent with previous reports (Akiyama et al., 2005; Murdiyarso et al., 2010). When the soil water content was below saturation, N<sub>2</sub>O emissions increase with soil moisture; however, N<sub>2</sub>O emissions gradually decreased with the soil saturation condition (Rudaz et al., 1999). A relatively high N<sub>2</sub>O peak was observed in the first two weeks of the wheat-growing season (Fig. 3), possibly because soil changes from flooded to drained conditions may have enhanced N<sub>2</sub>O release (Deng et al., 2012). Alternation of drainage and flooding may induce large amounts of N<sub>2</sub>O emissions, particularly in fertilized systems; this has commonly been shown in earlier studies (Wang et al., 2012; Xiong et al., 2007; Zou et al., 2005). The seasonal and annual rates of N<sub>2</sub>O emissions were significantly affected by the cultivation practices and years (Table 3). Compared with the FP plot, the ISSM-N2 scenario significantly decreased the seasonal N<sub>2</sub>O emissions in this study, which may have resulted from a reduction in the N fertilizer rate 366 (Table 1, Table 2). The total N<sub>2</sub>O emissions decreased by 7–38% and 26–42% in the rice and wheat seasons, respectively, when the conventional N management (300 kg N ha<sup>-1</sup> for rice 367 and 180 kg N ha<sup>-1</sup> per crop for wheat) changed to optimum N management (225-270 kg N 368 ha<sup>-1</sup> for rice and 135-162 kg N ha<sup>-1</sup> per crop for wheat). It is likely that more N<sub>2</sub>O was 369 370 emitted (Mosier et al., 2006) as a result of the additional N made available to the soil 371 microbes through N fertilizer application, which also probably contributed increased CH<sub>4</sub> 372 emissions (Banger et al., 2013). Strategies that can reduce N fertilization rates without 373 influencing crop yields can inevitably lower GHG emissions (Mosier et al., 2006). Nitrogen 374 leaching and volatilization are the important components of reactive N releases but not 375 included in the current GHG budget. 376 4.3 GWP and GHGI as affected by ISSM strategies The GWP in our study (10871–22711 kg CO<sub>2</sub> eq. ha<sup>-1</sup>) with the ISSM strategies was higher 377 than that in a double-cropping cereal rotation (1346–4684 kg CO<sub>2</sub> eq. ha<sup>-1</sup>) and a rice-wheat 378 379 annual rotation (290–4580 kg CO<sub>2</sub> eq. ha<sup>-1</sup>) reported by Huang et al. (2013b) and Yang et al. (2015), respectively. Dominant CH<sub>4</sub> emissions as well as additional CO<sub>2</sub> emissions due to the 380 381 use of machinery/equipment for irrigation and farm operations under the ISSM strategies may 382 increase the GWP more than in other cropping systems (emit more CO<sub>2</sub> equivalent emissions of 2439-5694 kg CO<sub>2</sub> eq. ha<sup>-1</sup> for agricultural management practices in the present study). 383 However, the current GWP was comparable to that of a double-rice cropping system 384 (13407-26066 kg CO<sub>2</sub> eq. ha<sup>-1</sup>) (Shang et al., 2011). The GHGIs, which ranged from 0.71 to 385 1.25 kg CO<sub>2</sub> eq. kg<sup>-1</sup> grain in this study, were slightly higher than previous estimates of 386 0.24-0.74 kg CO<sub>2</sub> eq. kg<sup>-1</sup> grain from rice paddies with midseason drainage and organic 387 manure incorporation (Qin et al., 2010; Li et al., 2006) but were lower than the DNDC model 388 estimates for continuous waterlogged paddies (3.22 kg CO<sub>2</sub> eq. kg<sup>-1</sup> grain) (Li et al., 2006). 389 390 Differences in GWP or GHGI were found in the cultivation patterns over the three rice-wheat 391 rotations (Table 5). The ISSM-N1 and ISSM-N2 scenarios with optimized ISSM strategies led 392 to a lower GWP than the FP by a certain extent, but there were not significant differences 393 among the FP, ISSM-N1 and ISSM-N2 plots (Table 5). Compared with the FP, the ISSM-N1

and ISSM-N2 scenarios significantly dramatically reduced the GHGI, which was mainly due

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(0.71 kg CO<sub>2</sub> eq. kg<sup>-1</sup> grain) was obtained under the ISSM-N2 scenario. This finding is consistent with the suggestion made by Burney et al. (2010), i.e., that the net effect of higher yields offsets emissions. It is well known that CH<sub>4</sub> emissions dominate the GWP in rice paddies (Ma et al., 2013; Shang et al., 2011). In comparison to the GWP (12371 kg CO<sub>2</sub> eq. ha<sup>-1</sup>yr<sup>-1</sup>) and GHGI (0.87 kg CO<sub>2</sub> eq. kg<sup>-1</sup> grain) of the FP, the ISSM-N3 and ISSM-N4 scenarios increased both the GWP and GHGI, mainly because these scenarios notably increased the CH<sub>4</sub> emissions compared with the FP, which resulted in relatively higher GWP (Table 5).

Agricultural management practices that change one type of GWP source/sink may also impact other sources/sinks and therefore change the GWP and GHGI (Mosier et al., 2006; Shang et al., 2011). Although the N-fertilizer plots, especially those with the incorporation of organic fertilizer, increased the annual CH<sub>4</sub> and N<sub>2</sub>O emissions, they increased the SOC sequestration in this cropping system, which is agreement with previous reports (Huang and Sun, 2006). This was mainlymay be due to the enhanced incorporation of rapeseed cake and enhanced below-ground crop residue associated with higher crop productivity (Ma et al., 2013). In the present study, the ISSM-N2 scenario with ISSM strategies decreased the CH<sub>4</sub> and N<sub>2</sub>O emissions as well as the energy consumption related to irrigation and the manufacture and transport of N fertilizer (depending on coal combustion), ultimately leading to a decrease in the GWP relative to the FP plot. Moreover, despite the lower N fertilizer input, the grain yield did not decline and the GHGI of the ISSM-N2 scenario was thus lower than of the FP plot, indicating less consumption of CO<sub>2</sub> equivalents per unit of grain produced. We demonstrate that high yield and agronomic NUE, together with low GWP, are not conflicting goals by optimizing ISSM strategies.

4.4 Main components of GWP and GHGI and implementation significance for the ISSM strategies

Determining the main components of the GWP and GHGI in specific cropping systems is very important for mitigating GHG emissions in the future, because the benefits of C sequestration would be negated by CH<sub>4</sub> and N<sub>2</sub>O emissions and the CO<sub>2</sub> equivalents released with the use of high N fertilizer application rates (Schlesinger, 2010). In the current study, the five main components of the CO<sub>2</sub> equivalents for the GWP were ranked in decreasing order of

importance as follows:  $CH_4$  emissions > agrochemical inputs of N fertilizer > farm operations related to irrigation > SOC sequestration >  $N_2O$  emissions (Table 5). In eachOf the two crops,  $CH_4$  and irrigation were important for rice, but less important for wheat, in which  $N_2O$  losses were expected to have a higher weight (Supplementary resource 2). Methane emissions, the most important component of GWP in this typical rice-wheat rotation system, could be further mitigated by some other strategies, such as reasonable irrigation (Zou et al., 2005; Wang et al., 2012).

Although N fertilizer application increased SOC sequestration when it was applied with rapeseed cake manure, this benefit was consistently overshadowed, on a CO<sub>2</sub> equivalent basis, by the increases in CH<sub>4</sub> and N<sub>2</sub>O emissions (Table 5). Similar results have been reported, i.e., GHG emissions substantially offset SOC increases (Six et al., 2004). It is possible that the realization of reducing the GWP and GHGI in China should focus on increasing the SOC and simultaneously decreasing the CO<sub>2</sub> equivalents from CH<sub>4</sub> emissions and N fertilizer inputs. Several studies reported possible methods for these types of mitigation strategies, such as optimizing the chemical fertilizer application amount and rate (Ju et al., 2011), the amount of water used for irrigation (Gao et al., 2015), and the timing and rate of N using the in-season N management approach, as well as improving the N fertilizer manufacturing technologies (Zhang et al., 2013), and using nitrification inhibitors or polymer-coated controlled-release fertilizers (Hu et al., 2013).

China is a rapidly developing country that faces the dual challenge of substantially increasing grain yields and at the same time\_as\_reducing the very substantial environmental impacts of intensive agriculture (Chen et al., 2011). We used the ISSM strategies to develop a rice production system that achieved mean yields of 10.63 t ha<sup>-1</sup> (an increment of almost 24%) and an agronomic NUE of 13.20 kg grain kg N<sup>-1</sup> (an increment of 43%) in long-term field experiments compared with current farmers' practices. The ISSM redesigned the whole production system only for the rice crop based on the local environment and drawing on appropriate fertilizer varieties and application ratios, crop densities and an advanced water regime management. If the ISSM strategies were also developed for the rotated wheat crop, the overall performance of the whole rice-wheat system would be much improved, with further increases in yield and reductions in the GWP and GHGI. We conclude that the ISSM

strategies are promising, particularly the ISSM-N2 scenario, which is the most favorable to realize higher yields with lower environmental impact. The proposed ISSM strategies can provide substantial benefits to intensive agricultural systems and can be applied feasibly using current technologies.

### **5 Conclusions**

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Reasonable agricultural management practices are the key to reducing GHG emissions from agricultural ecosystems. This study provided an insight into the complete GHG emission accounting of the GWP and GHGI affected by different ISSM scenarios. After a fivethree-year field experiment, we found that the CH<sub>4</sub> emissions, production of N fertilizer, irrigation, SOC sequestration and N2O fluxes were the main components of the GWP in a typical rice-wheat rotation system. In contrast with the FP, ISSM-N1 and ISSM-N2 significantly reduced the GHGI, though they resulted in similar GWPs, and ISSM-N3 and ISSM-N4 remarkably increased the GWP and GHGI. By adopting the ISSM-N2 strategy, the conventional N application rate was reduced by 10% while the rice yield was significantly increased by 16%, the NUE was improved by 67% and the GHGI was lowered by 23%. ISSM scenarios could be adopted for both food security and environmental protection with specific targets. We propose that the ISSM-N2 scenario is the most appropriate management strategy (10% reduction of N input, no rapeseed manure and higher plant density) for realizing higher yields and NUE, together with some potential to reduce GHGI by integrated soil-crop management. For simultaneously mitigating GHG emissions, further research on integrated soil-crop system managements is required particularly for mitigating CH<sub>4</sub> emissions in sustainable rice agriculture. Acknowledgments We sincerely appreciate two anonymous reviewers for their critical and valuable comments to help improve this manuscript. This work was jointly supported by the National Science Foundation of China (41171238, 41471192), the Special Fund for Agro-Scientific Research in the Public Interest (201503106) and the Ministry of Science and Technology (2013BAD11B01).

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**Table 1**The establishment of different treatments for the annual rice-wheat rotations during the 2011–2014 cycle.

Scenario	$NN^a$	FP	ISSM-N1	ISSM-N2	ISSM-N3	ISSM-N4						
	Rice-growing season											
Chemical fertilizer application rate (N:P <sub>2</sub> O <sub>5</sub> :K <sub>2</sub> O:Na <sub>2</sub> SiO <sub>3</sub> :ZnSO <sub>4</sub> , kg ha <sup>-1</sup> )	0:90:120:0:0	300:90:120:0:0	225:90:120:0:0	270:90:120:0:0	300:108:144:225:15	375:126:180:225:15						
Split N application ratio		6:2:0:2	5:1:2:2	5:1:2:2	5:1:2:2	5:1:2:2						
Rapeseed cake manure (t ha <sup>-1</sup> )	0	0	0	0	2.25°	2.25						
Water regime	F-D-F-M <sup>b</sup>	F-D-F-M	F-D-F-M	F-D-F-M	F-D-F-M	F-D-F-M						
Planting density (cm)	20×20	20×20	20×15	20×15	20×15	20×20						
			Whe	at-growing season	1							
Chemical fertilizer application rate (N:P <sub>2</sub> O <sub>5</sub> :K <sub>2</sub> O, kg ha <sup>-1</sup> )	0:90:180	180:90:180	135:90:180	162:90:180	180:108:216	225:126:270						
Split N application ratio		6:1:3	6:1:3	6:1:3	6:1:3	6:1:3						
Seed sowing density (kg ha <sup>-1</sup> )	180	180	180	180	180	180						

<sup>&</sup>lt;sup>a</sup>NN, no N application; FP, farmers' practice; The four integrated soil-crop system management (ISSM) practices at different nitrogen application rates relative to the FP rate of 300 kg N ha<sup>-1</sup> for the rice crop and 180 kg N ha<sup>-1</sup> for the wheat crop, namely, ISSM-N1 (25% reduction), ISSM-N2 (10% reduction), ISSM-N3 (FP rate) and ISSM-N4 (25% increase). Urea, calcium biphosphate and potassium chloride were used as N, P and K fertilizer respectively.

<sup>&</sup>lt;sup>b</sup>F-D-F-M, flooding-midseason drainage-re-flooding-moist irrigation.

<sup>°112.5</sup> kg N ha<sup>-1</sup> in the form of rapeseed cake was applied as basal fertilizer and included in the total N rate for calculating agronomic NUE.

 $\begin{tabular}{ll} \textbf{Table 2} \\ \textbf{Seasonal $CH_4$ and $N_2O$ emissions, and yields during rice and wheat cropping seasons in the three} \\ \textbf{cycles of 2011-2014}. \\ \end{tabular}$ 

		Rice season			Wheat season	
Treatment	CH <sub>4</sub>	N <sub>2</sub> O	Yield	CH <sub>4</sub>	N <sub>2</sub> O	Yield
	(kg C ha <sup>-1</sup> )	$(kg \ N \ ha^{-1})$	$(t ha^{-1})$	$(kg C ha^{-1})$	(kg N ha <sup>-1</sup> )	$(t ha^{-1})$
2011						
NN	153±10.8c	0.03±0.05c	5.85±0.08f	- 0.48±0.63a	$0.45\pm0.09d$	1.74±0.18d
FP	266±25.3b	0.11±0.08c	8.38±0.35e	- 0.48±1.86a	1.43±0.19b	5.67±0.20b
ISSM-N1	212±30.3bc	0.08±0.03c	9.27±0.26d	$0.78\pm0.97a$	0.65±0.11cd	5.05±0.16c
ISSM-N2	220±32.5bc	0.17±0.11bc	9.79±0.44c	$2.25\pm2.07a$	$0.80 \pm 0.06c$	$5.71\pm0.18b$
ISSM-N3	518±58.9a	0.38±0.15ab	$10.81 \pm 0.26b$	0.04±3.23a	1.40±0.10b	5.31±0.26bc
ISSM-N4	561±50.9a	0.37±0.07a	11.76±0.24a	- 0.09±1.40a	1.93±0.09a	6.15±0.15a
2012						
NN	149±25.8d	0.13±0.10c	$5.80\pm0.22f$	- 4.32±7.29a	0.65±0.09d	1.73±0.11c
FP	239±34.5c	0.33±0.11bc	8.72±0.62e	4.85±10.30a	2.13±0.43ab	5.64±0.34ab
ISSM-N1	226±30.4cd	0.27±0.07bc	9.43±0.34d	1.46±6.38a	1.39±0.14c	4.94±0.38b
ISSM-N2	228±32.6cd	0.38±0.29bc	9.99±0.50c	- 1.02±0.84a	1.77±0.38bc	5.78±0.59ab
ISSM-N3	431±26.8b	0.52±0.16ab	10.92±0.61b	2.45±8.35a	2.19±0.24ab	5.39±0.39ab
ISSM-N4	536±58.7a	0.78±0.13a	12.24±0.60a	5.91±6.18a	2.61±0.42a	6.10±0.49a
2013						
NN	101±39.2b	0.16±0.09b	5.84±0.15f	- 1.45±1.34a	0.35±0.06c	1.80±0.03c
FP	141±25.2b	$0.43\pm0.39ab$	8.67±0.26e	- 3.70±1.76a	$0.80 \pm 0.20 ab$	5.70±0.30ab
ISSM-N1	135±15.7b	0.19±0.16ab	9.66±0.29d	- 1.00±1.61a	0.49±0.16bc	5.15±0.20b
ISSM-N2	129±32.2b	0.26±0.13ab	10.15±0.07c	- 0.79±1.60a	0.69±0.24abc	5.80±0.18ab
ISSM-N3	256±45.6a	0.59±0.42ab	11.14±0.10b	- 0.62±1.14a	$0.71 \pm 0.10ab$	5.51±0.33ab
ISSM-N4	304±22.3a	0.74±0.40a	12.34±0.16a	$0.55\pm1.68a$	1.02±0.11a	6.19±0.63a
Average 20	11-2013 <sup>a</sup>					
$NN^b$	135±19.6d	0.11±0.05c	5.83±0.04f	- 2.08±1.89a	$0.48 \pm 0.07 d$	1.75±0.04d
$FP^b$	215±19.9c	0.29±0.13bc	8.59±0.25e	0.22±3.96a	1.45±0.24b	5.67±0.16b
ISSM-N1 <sup>b</sup>	191±19.2c	0.18±0.06c	9.45±0.18d	$0.42\pm2.77a$	$0.84 \pm 0.08c$	5.04±0.08c
ISSM-N2 <sup>b</sup>	192±11.6c	0.27±0.12bc	9.98±0.25c	0.15±0.58a	1.08±0.12c	5.76±0.22ab
ISSM-N3 <sup>b</sup>	402±23.8b	0.50±0.16ab	10.95±0.13b	0.63±3.51a	1.43±0.05b	5.40±0.16bc
ISSM-N4 <sup>b</sup>	467±39.2a	0.68±0.15a	12.11±0.28a	2.12±2.57a	1.85±0.16a	6.14±0.35a

<sup>a</sup>Mean  $\pm$  SD, different lower case letters within the same column for each item indicate significant differences at P<0.05 according to Tukey's multiple range test.

<sup>&</sup>lt;sup>b</sup>See Table 1 for treatment codes.

Table 3 Repeated-measures analysis of variance (MANOVA) for the effects of cultivation patterns (P) and cropping year (Y) on mean  $CH_4$  and  $N_2O$  emissions, and mean rice and wheat grain yields in the 2011-2014 cycle.

Crop season	Source	df	CH <sub>4</sub>	$N_2O$	Yield
			(kg C ha <sup>-1</sup> )	(kg N ha <sup>-1</sup> )	(t ha <sup>-1</sup> )
Rice	Between subjects				
	P	5	35.3***	3.71***	123***
	Within subjects				
	Y	2	20.7***	0.88**	1.15**
	$P \times Y$	10	6.73***	0.15	0.37
Wheat	Between subjects				
	P	5	0.26	14.8***	76.3***
	Within subjects				
	Y	2	0.55*	15.1***	0.08
	$P \times Y$	10	0.83	4.39***	0.05
Rice-Wheat	Between subjects				
	P	5	37.2***	24.2***	153***
	Within subjects				
	Y	2	20.5***	5.83***	0.70*
	$P \times Y$	10	6.50***	1.11	0.17

df – degrees of freedom, \* P < 0.05, \*\*\* P < 0.01, and \*\*\*\*P < 0.001 represent significant at the 0.05, 0.01 and 0.001 probability level, respectively.

Table 4

Agricultural management practices for chemical inputs and farm operations and contributions to carbon dioxide equivalents (kg CO<sub>2</sub> eq. ha<sup>-1</sup>yr<sup>-1</sup>) in the annual rice-wheat rotations from 2011 to 2014 (chemical inputs and farm operations used in each year were similar except for irrigation water).

Treatment	Chemical inputs (kg ha <sup>-1</sup> ) <sup>a</sup>								Farm operations <del>(kg ha ' )</del> c							
	N	P	K	Si	Zn	Herbicide	Insecticide	Fungicide	Irri	Irrigation (cm) <sup>b</sup>		Tillage and <u>raking</u>	Crop planting	<u>Farm</u> <u>manure</u>	Crop harvest	ThreshingFa machinery
									2011	2012	2013	(kg diesel ha <sup>-1</sup> ) <del>raking</del>	(event)	(kg ha <sup>-1</sup> )	(kg diesel ha <sup>-1</sup> )	(kw•h ha <sup>-1</sup> ) <del>product</del>
$NN^d$	0	180	300	0	0	2	18	4	75	80	80	37	2	0	11	135
FP	480	180	300	0	0	2	20	4.4	75	80	80	37	2	0	11	147
ISSM-N1	360	180	300	0	0	2	20	4.4	50	65	55	37	2	0	11	139
ISSM-N2	432	180	300	0	0	2	20	4.4	50	65	55	37	2	0	11	177
ISSM-N3	480	216	360	225	15	2	27	6	50	65	55	37	2	2250	11	177
ISSM-N4	600	252	450	225	15	2	41	9	50	65	55	37	2	2250	11	275
			C	Chemic	al inp	outs (Ei) (kg C	${\rm CO_2} \ {\rm eq.} \ {\rm ha}^{-1})$					Farm op	erations (Eo)	(kg CO <sub>2</sub> eq.	ha <sup>-1</sup> )	
NN	0	132	165	0	0	46	338	53	1419	1514	1514	127	23	0	37	36
FP	2288	132	165	0	0	46	375	59	1419	1514	1514	127	23	0	37	39
ISSM-N1	1716	132	165	0	0	46	375	59	946	1230	1041	127	23	0	37	37
ISSM-N2	2059	132	165	0	0	46	375	59	946	1230	1041	127	23	0	37	47
ISSM-N3	2288	158	198	58	6	46	506	79	946	1230	1041	127	23	62	37	47
ISSM-N4	2860	185	248	58	6	46	768	129	946	1230	1041	127	23	62	37	73

<sup>&</sup>lt;sup>a</sup>The carbon emission coefficients were 1.3,0.2,0.15, 6.3, 5.1 and 3.9 C cost (kg C eq. kg<sup>-1</sup> active ingredient) per applied nitrogen fertilizer, phosphorus, potassium, herbicide, insecticide and fungicide, respectively, as referred to in Lal (2004). We collected data specific to China's fertilizer manufacture and consumption, and then estimated carbon emissions coefficients were 0.07 and 0.1 C cost (kg C eq. kg<sup>-1</sup> active ingredient) per applied Si and Zn fertilizer, respectively.

<sup>&</sup>lt;sup>b</sup>The carbon emission coefficient for irrigation was 5.16 C cost (kg C eq. cm<sup>-1</sup> ha<sup>-1</sup>) as referred to in Lal (2004).

<sup>&</sup>lt;sup>c</sup>The carbon emission coefficients were 0.94 <u>C cost (kg C eq. kg<sup>-1</sup> diesel) for tillage, raking and harvesting, 3.2 <u>C cost (kg C eq. event<sup>-1</sup>) for crop planting, for per farm manure application was 0.0075 <u>C cost (kg C eq. kg<sup>-1</sup>) for farm manure application, 0.94</u> and for threshing was 0.0725 <u>C cost (kg kg</u> C eq. (kw•h)<sup>-1</sup> kg<sup>-1</sup> active ingredient) for threshing, erop planting, per farm</u></u>

manure application, harvesting, spraying and threshing, respectively, as referred to in Lal (2004).

<sup>d</sup>See Table 1 for treatment codes.

Table 5

Mean global warming potential (GWP) and greenhouse gas intensity (GHGI) over the three rice season, wheat season and annual cycles of the 2011rice season–2014wheat season.

Treatment	CH <sub>4</sub>	$N_2O$	Ei	Eo	SOCSR	$GWP^a$	Grain yield	GHGI <sup>b</sup>		
			kg CO <sub>2</sub> ec	q. ha <sup>-1</sup> yr <sup>-1</sup>			t ha <sup>-1</sup> yr <sup>-1</sup>	kg CO <sub>2</sub> eq. t <sup>-1</sup> grain		
Rice season										
NN	5026±733d	44±20c	424	1601	-396±164c	7492±706d	$5.83 \pm 0.04 f$	1285±123b		
FP	8035±742c	121±53bc	1859	1603	585±198ab	11032±555c	8.59±0.25e	1285±68b		
ISSM-N1	7132±716c	75±24c	1502	1191	246±218b	9654±800c	$9.45 \pm 0.18d$	1021±81c		
ISSM-N2	7186±434c	112±49bc	1716	1198	355±97ab	9858±484c	9.98±0.25c	989±67c		
ISSM-N3	15005±888b	208±66ab	2037	1260	691±252a	17818±786b	10.95±0.13b	1626±54a		
ISSM-N4	17427±1463a	284±60a	2626	1280	773±174a	20844±1452a	12.11±0.28a	1720±108a		
Wheat seaso	Wheat season									
NN	-78±71a	201±28d	310	104	-396±164c	934±214b	1.75±0.04d	533±125a		
FP	8±148a	605±99b	1206	105	585±198ab	1339±129b	5.67±0.16b	236±21b		
ISSM-N1	16±103a	351±32c	991	105	246±218b	1217±342b	5.04±0.08c	241±68b		
ISSM-N2	6±22a	451±49c	1120	108	355±97ab	1329±109b	$5.76\pm0.22ab$	231±26b		
ISSM-N3	23±131a	598±20b	1302	108	691±252a	1340±290b	5.40±0.16bc	247±48b		
ISSM-N4	79±96a	772±66a	1674	114	773±174a	1867±175a	$6.14\pm0.35a$	305±33b		
Rice-wheat	rotation									
$NN^d$	$4948 \pm 704 d^{c}$	246±26d	734	1705	-792±327c	8425±711d	7.58±0.04d	1111±94b		
FP	8043±858c	725±49b	3065	1708	1170±396ab	12371±583c	14.26±0.36c	868±29c		
ISSM-N1	7141±709c	426±55c	2493	1296	491±435b	10871±990c	14.50±0.14c	750±68d		
ISSM-N2	7192±424c	563±86c	2836	1306	709±193ab	11187±552c	15.74±0.44b	712±52d		
ISSM-N3	15028±833b	806±77b	3339	1368	1383±503a	19158±761b	16.36±0.18b	1171±37ab		
ISSM-N4	17506±1396a	1056±58a	4300	1394	1545±348a	22711±1438a	18.26±0.46a	1245±93a		

 $<sup>^{</sup>a}$ GWP (kg CO<sub>2</sub> eq. ha $^{-1}$ yr $^{-1}$ ) = 28 × CH<sub>4</sub> +265 × N<sub>2</sub>O + Ei +Eo - 44/12 × SOCSR, Ei (agrochemical inputs), Eo (farm operations),

SOCSR (SOC sequestration rate) is divided by 2 to roughly estimate the GWP from rice and wheat season, respectively. All other items were actually measured for each season.

<sup>c</sup>Different lower case letters within the same column for each item indicate significant differences at *P*<0.05 based on Tukey's multiple range tests.

<sup>&</sup>lt;sup>b</sup>GHGI (kg CO<sub>2</sub> eq. t<sup>-1</sup> grain) = GWP/grain yields

<sup>&</sup>lt;sup>d</sup>See Table 1 for treatment codes.

Fig 1 Rice and wheat agronomic nitrogen use efficiency (NUE) in 2011–2014 in Changshu, China. Different letters indicate a significant difference between treatments (p<0.05). See Table 1 for treatment codes.

Fig 2 Seasonal variation of methane (CH<sub>4</sub>) fluxes from the rice-wheat rotation cropping systems from 2011 to 2014. The black and gray part in figure separates different grain growth periods. See Table 1 for treatment codes. The solid arrows indicate fertilization.

Fig 3 Seasonal variation of nitrous oxide ( $N_2O$ ) fluxes from rice-wheat rotation cropping systems in three annual cycles over the period 2011–2014. The black and gray part in the figure separates different growth periods. See Table 1 for treatment codes. The solid arrows indicate fertilization.

Supplementary resource 2 Agricultural management practices for chemical inputs and farm operations in the rice and wheat cropping seasons.

								]	Rice season	1						
				Chen	nical i	nputs (kg h	$a^{-1}$ ) <sup>a</sup>		Farm operations <sup>b</sup>							
									Irrigation	Tillage	Crop	Farm	Crop	Threshing <sup>c</sup>		
	N	P	K	Si	Zn	Herbicide	Insecticide	Fungicide			planting	manure	harvest			
Treatment									(cm)	(kg diesel ha <sup>-1</sup>	(event)	(kg ha <sup>-1</sup> )	) (kg diesel ha	(kw•h ha <sup>-1</sup> )		
NN	0	90	120	0	0	1	13	2	78	20	1	0	6	74		
FP	300	90	120	0	0	1	13	2.4	78	20	1	0	6	80		
ISSM-N1	225	90	120	0	0	1	13	2.4	57	20	1	0	6	74		
ISSM-N2	270	90	120	0	0	1	13	2.4	57	20	1	0	6	100		
ISSM-N3	300	108	144	225	15	1	17	3.5	57	20	1	2250	6	100		
ISSM-N4	375	126	180	225	15	1	26	5	57	20	1	2250	6	175		
								V	Vheat seaso	n						
NN	0	90	180	0	0	1	5	2	0	17	1	0	5	61		
FP	180	90	180	0	0	1	7	2	0	17	1	0	5	67		
ISSM-N1	135	90	180	0	0	1	7	2	0	17	1	0	5	65		
ISSM-N2	162	90	180	0	0	1	7	2	0	17	1	0	5	77		
ISSM-N3	180	108	216	0	0	1	10	2.5	0	17	1	0	5	77		
ISSM-N4	225	126	270	0	0	1	15	4	0	17	1	0	5	100		

<sup>&</sup>lt;sup>a</sup>There was no machinery used for fertilizer application.

<sup>&</sup>lt;sup>b</sup>Tillage and crop harvest, crop planting, and threshing were calculated by diesel fuel (kg ha<sup>-1</sup>), event and electricty (kw•h ha<sup>-1</sup>), respectively.

<sup>&</sup>lt;sup>c</sup>Electricity energy is calculated according to the power and working hours. The power of thresher is 15 kilowatt in this experiment.