Dear Reviewer#1, Thank you very much for your critical comments and great support! Please see the attached point-by-point answers and the manuscript with tracking system for your further evaluation.

Sincerely yours,

Zhengqin (on behalf of all authors)

Referee #1

GENERAL COMMENTS The authors present an interesting and complete assessment on Global Warming Potential (GWP) and greenhouse gas intensity (GHGI) during three years in a rice-wheat rotation. The number of crop seasons, as well as the complete overview the sustainability of the agro-ecosystem (soil GHG emissions, SOC, CO2 equivalents from inputs and operations, and crop yields) are, from my point of view, the main strengths of the this study, which fits well into the scope of the journal. Conversely, the manuscript requires additional details and explanation before it can be considered for publication. Moreover, I do not understand why the authors did not set some variables (e.g. Zn fertilization -which has been reported to influence crop yields and GHG emissions- plant density, water management...). That would have simplified the discussion and maybe would have allowed obtaining some conclusions about management techniques (and not only about the overall scenarios) and the possibilities of combining scenarios. The authors should also improve the Materials and Methods section, explaining much better the GWP calculations and other issues of major interest. The conclusions are adequately presented: since each scenario is a combination of several management techniques, the authors cannot recommend any single practice, only the full scenario. Conversely, ALL the management factors that could have influence the measured variables (yields, GHG fluxes, GWP) should be briefly discussed.

A: Thank you very much for your great support and critical comments. Those comments are all valuable and very helpful for revising and improving our paper, as well as further important guidance for our researches. We have made corrections which we hope to meet with approval. Please see the following point-by-point answers.

1. Yes, all of these variables such as Zn fertilization, plant density, and water management affect crop yields and GHG emissions. We are sorry that we did not set them as separate variables and just integrated them to realize our goal for better yield and NUE. According to your comments, we added some information for better understanding in discussion, such as on Page 10, Lines 262-265 and 270-275 for yield, Page 12, Lines 306-308 for GHG emissions.

2. It is really true that we update all possible components for calculating GWP as both of you Referee suggested. So, we put them in a better way on Page 6, Lines 149-173.

3. In conclusion, the ISSM scenarios could be adopted for both food security and environmental protection. We discussed scenarios in detail emphasizing the main components. Revised accordingly Page 14, Lines 385-388 and Page 16, Lines 430-432.

SPECIFIC COMMENTS

Abstract, lines 19-23. It seems that treatments only differ in N rate, and that is not true. Please indicate briefly other parameters that were different between treatments.

A: Thank you for your comment. Revised accordingly Page 2, Lines 18-20.

Lines 82-84. Include, if available, the information on clay, silt and sand contents and soil texture. A: Thank you for your comment. Revised accordingly Page 4, Lines 90-91.

Lines 84-86. Please indicate the annual precipitation and mean temperature in each campaign.

Were "normal years" with regard to these parameters, compared to the average values (15.5 $^{\circ}$ C and 1038 mm)? In any case, I think that this information fits better in the results section.

A: Yes, thank you. Revised accordingly Page 4, Lines 88-89.

Lines 88-98. What was the N source applied? What were the dates (or phenological stages) of N application in rice -4 split applications in some treatments-? What about the wheat? When was the N applied?

A: Urea was used as N fertilizer and 20 kg N ha⁻¹ in the form of rapeseed cake fertilizer was applied as N fertilizer for N3 and N4 treatments in this study. Revised accordingly Page 5, Lines 112-119.

Please indicate that Zn and Na2SiO3 fertilization, plant density and the use of an organic amendment were also management techniques for improving rice yield and NUE.

A: Thank you very much for your comment! According to your comments, we added some information for better understanding in discussion.

Did you consider the N supplied through rapeseed cake manure for NUE calculations (you should!)? In any case, please indicate in this section the composition of this residue, at least the C and N contents and C:N ratio.

A: We are sorry for the inconvenience. We recalculated the NUE according to your comment.. Revised accordingly Page 5, Line 102, Page 8, Lines 196-199 and Fig. 1.

Lines 99-104. What amount of irrigation water was applied during the rice growing season in each campaign? Was it different between scenarios (as could be deduced from line 329)?

A: The amount of irrigation water applied during the rice growing season in each campaign was as shown in Table 4. It was different between ISSM scenarios and FP and NN.

Please describe briefly the residue management at the end of rice and wheat growing season.

A: The rice and wheat straws were removed out of the field for all the treatments in this study. Revised accordingly Page 5, Lines 119-120.

Line 116. To my knowledge, it is not possible to measure N2O with a flame ionization detector. Was not your GC equipped with an ECD detector? Please, clarify this crucial point.

A: We are sorry for the inconvenience. All of the gas samples were analyzed using a modified gas chromatograph (Agilent 7890A, Shanghai, China) equipped with two detectors. N₂O was detected by an electron capture detector (ECD). Argon-methane (5%) was used as the carrier gas at a flow rate of 40 ml min⁻¹ for the N₂O analysis. The temperatures of the column and the ECD were maintained at 40 °C and 300 °C, respectively. Revised accordingly Page 6, Lines 132-137.

Line 125. Can you reference this equation?

A: Yes. Revised accordingly Page 6, Line 142.

Lines 131-149. This section needs to be significantly improved. I recommend the authors to summarize the information in a DETAILED Table, indicating for each variable that is included in GWP calculations: Amount/number of labors/inputs, unit cost (kg CO2 equivalents) and a reference.

A: Thank you very much for your comment! Please see the Table 4 for the detailed information.

The information on fertilizer rates is enough, but you should indicate the number of farm operations (labor passes, seeding, fertilizing, harvesting, irrigation –and amount of water applied-) in each treatment. Were the ZnSO4 and Na2SiO3 considered for the GWP? (They should!)

A: Thank you for your comment. We added a new Table 4 for understanding the Eo and Ei components. The $ZnSO_4$ and Na_2SiO_3 were also considered for the GWP. There must have other

uncertainties in selecting parameters. This is only preliminary evaluation within our understanding. Thank you very much for your great support!

What does the Ei involve? Only inputs application? Application + manufacture and transport? A: Ei involve application + manufacture and transport.

What are the CO2 equivalents for N2O and CH4 emissions? Please, indicate (with a reference). A: Thank you for your comment. Revised accordingly Page 6, Line 152.

Lines 151-157. Were the normal distribution and variance uniformity checked? Did you use any non-parametric test for non-normally distributed data? If affirmative, please indicate how.

A: We have made the normal distribution and variance uniformity check. All data were conformed.

Line 167-168. I don't know what you meant here. According to Table 2, significant increases were observed... If this statement is referred to average wheat-rice values, please add "data not shown" in brackets.

A: Thank you for your comment. This statement was meant that significant increase was observed only in N4 scenario in wheat grain yields.

NUE: See my comment on lines 88-98 for NUE calculations in N3 and N4 scenarios (with rapeseed cake manure). Correct NUE for these cultivation patterns if the N from rapeseed was not considered.

A: We are sorry for the inconvenience. 20 kg N ha⁻¹ in the form of rapeseed cake fertilizer was applied as basal fertilizer and should be included in the total N rate. Revised accordingly Page 8, Lines 196-199 and Fig. 1.

Line 175-176. Please include this sentence "current ISSM strategy was only designed for rice production, not wheat production" in Materials and Methods.

A: Thank you for your comment. Revised accordingly Page 5, Lines 100-101.

Line 187-188. Avoid as possible the subjective statements (e.g. "due to the combined application of inorganic and organic fertilizers") in the results section. This sentence (from my point of view) fits better in the discussion.

A: Thank you for your comment. Deleted accordingly Page 9, Line 215.

Lines 189-196. The description of the N2O evolution should be improved. You should indicate how many peaks or increments (if any) were reported in each crop, and if they were reported after fertilization events. Comparing emissions (CH4 and N2O) between crops (wheat versus rice) would be interesting.

A: Thank you for your comment. Revised accordingly Page 9, Lines 216-218.

"Correlations between seasonal cumulative N2O emissions and fertilizer N application rates were also calculated": that should be indicated in Materials and Methods (Statistical Analysis section, in which you only explain that "linear relationships were determined"). About this regression, the N rate was not the only variable that could have influenced N2O emissions, since other variables were not fixed. Moreover, the N rate in N3 and N4 is not correct (you have not considered rapeseed manure). Therefore, I recommend removing this figure.

A: Thank you for your comment. Deleted accordingly.

"Relative to the FP plot, the N1 and N2 scenarios decreased the annual N2O emissions by an average of 41% and 22%, while the N4 scenario significantly increased it by 46% although there was no significant difference between N3 and FP plots (P < 0.05)". This sentence is so confusing for me. Is this statement referred to wheat? If affirmative, please indicate and check the

percentages of abatement/enhancement (looking at Table 2, average 2011-2013, I think there are some mistakes). You should describe also the results for the rice (in spite of the lack of significant differences between fertilized treatments). If these percentages are based on information of Table 2, please refer to this Table.

A: Sorry for the inconvenience. This statement referred to the annual N_2O emission rates including the results for both rice and wheat. Revised accordingly Page 9, Lines 222-323.

"With respect to the N application effect, the annual cumulative N2O emissions in all four ISSM scenarios were significantly higher than in NN (P < 0.05)". In what crop? In wheat? Please indicate, because this is not observed for rice (Table 2, Average 2011-2013 fluxes).

A: Sorry for the inconvenience. The annual cumulative N_2O emissions indicate the average rates in rice-wheat rotations of 2011–2014.

Line 202-203. "Although N fertilizer increased annual CH4 and N2O emissions, they also increased SOC sequestration in this cropping system". Please change "N fertilizer" by "fertilized treatments". The idea that N fertilization can increase SOC sequestration is speculative, particularly in the Results section.

A: Thank you for your comment. Revised accordingly Page 9, Lines 233.

Lines 206-207: "CO2 equivalents from machinery used for Ei (2449–4192 CO2-eq ha–1 yr–1) were higher than Eo (1285–1697 CO2-eq ha–1 yr–1) of the fertilized plots". The word "machinery" is confusing form me since you explain (in lines 123-133) that the Ei component accounted the CO2 emissions from agrochemical inputs.

A: Sorry for the inconvenience. Revised accordingly Page 9, Lines 238.

Lines 211-212: "Consequently, the lowest NGWP was achieved under the N1 scenario for the ISSM." The NGWP in N1, N2 and FP was statistically similar. Please remove or correct this sentence.

A: Thank you for your comment. Deleted accordingly Page 9, Lines 243.

General comments on Discussion section: the Discussion should be significantly improved. First of all, the document (and particularly this section) should be reviewed by a native speaker. As explained above, each scenario is a combination of several management techniques, and even though authors cannot recommend any single practice (only the full scenario), ALL the management factors that could have influence the measured variables (yields, GHG fluxes, GWP) should be discussed:

A: Thank you for your comment. Revised accordingly throughout the discussion. We also asked AJE for re-editing the texts. Thank you for your patience. We added references (Kabata-Pendias and Mukherjee, 2007; Hossain et al., 2008) in the text.

• Fertilization: Not only different N rates were tried, but also different P, K, silicon and Zn. This fact may have directly affected crop yields, and also N2O emissions indirectly (i.e. the more a crop grows, the more N is uptaken and the less N is likely to be lost as N2O). Particularly, Zn has been reported to influence CH4 and N2O emissions (Glass and Orphan, 2012) and yields in some crops, e.g. rice (Hossain et al., 2008). The effects of silicon in rice physiology have also been described (Kabata-Pendias and Mukherjee, 2007).

Hossain, M. A., Jahiruddin, M., Islam, M. R., & Mian, M. H. (2008). The requirement of zinc for improvement of crop yield and mineral nutrition in the maize–mungbean–rice system. Plant and soil, 306(1-2), 13-22.

Glass, J. B., & Orphan, V. J. (2012). Trace metal requirements for microbial enzymes involved in

the production and consumption of methane and nitrous oxide. Frontiers in microbiology, 3. Kabata-Pendias, A., & Mukherjee, A. B. (2007). Trace elements from soil to human. Springer Science & Business Media.

• The rapeseed cake manure is also an important variable. This organic amendment can substantially modify CH4 and N2O fluxes thorough the release of C and N. You should indicate the composition of this residue (C and N contents) and discuss better its influence on GHG emissions (Thangarajan et al., 2013) and C sequestration. Moreover, the residues from Brassicaceae have been reported to inhibit nitrification, thus affecting N2O losses (Subbarao et al., 2015).

A: Thank you for your comment. Revised accordingly throughout the discussion. We added reference (Thangarajan et al., 2013) in the text.

Thangarajan, R., Bolan, N. S., Tian, G., Naidu, R., & Kunhikrishnan, A. (2013). Role of organic amendment application on greenhouse gas emission from soil. Science of the Total Environment, 465, 72-96.

Subbarao, G. V., Yoshihashi, T., Worthington, M., Nakahara, K., Ando, Y., Sahrawat, K. L., ... & Braun, H. J. (2015). Suppression of soil nitrification by plants. Plant Science, 233, 155-164.

• Plant density, split N application ratio and water regime were not the same in all treatments (I do not know why). Could these changes have modified GHG emissions and crop yields?

For instance, in lines 230-234, you consider that the changes in rice yields in N1 and N2 treatments as opposed to FP are due to changes in N fertilization rate. But the crop density and split ratio were different, and that could have also affected yields. In the reference that you provide (Peng et al., 2006) only the N rate is changed between treatments.

A: Thank you for your comment. Revised accordingly Page 10, Lines 262-265.

As indicated above, the N supplied thorough the manure should be taken into account for NUE calculations. Therefore, the NUE should be re-calculated and re-discussed.

A: Thank you for your comment. Revised accordingly Page 8, Lines 196-199 and Fig. 1.

Lines 239-240: "Possible explanations can be that organic fertilizer supplemented with adequate nutrients in combination with improved rice yield and efficient control of pests and diseases". As indicated above, many factors could have contributed to this higher yields and must be discussed (i.e. why the organic amendment increase yields? Maybe due to the higher supply of nutrients that you have not considered)... But what about the efficient control of pest and diseases? Were not efficiently controlled in the rest of treatments? This point needs to be clarified.

A: Thank you for your comment. Revised accordingly throughout the discussion.

Lines 248-252. Please split and/or rewrite this sentence for better understanding and be careful with the variables that were not measured (e.g. leaching and volatilization).

A: Thank you for your comment. Revised accordingly Page 11, Lines 282-285.

Line 277: add a reference for this statement.

A: Thank you for your comment. Revised accordingly Page 12, Lines 318.

Lines 278-279. Likely, the flooded conditions during rice season led to the reduction of N2O to N2 (complete denitrification).

A: Thank you for your comment.

Lines 280-282: add a reference for this statement.

A: Thank you for your comment. Revised accordingly Page 12, Lines 324.

Lines 286-287: as explained above, I recommend removing this correlation analysis.

A: Thank you for your comment. Deleted accordingly Page 12, Lines 328.

Lines 325-327: why the fertilized treatments could have increased SOC sequestration? That should be briefly discussed (maybe that could be related to residue management, which you should also explain).

A: Revised accordingly Page 14, Lines 368-369.

Line 343-344: I suggest adding a figure or table indicating the relative weight of each component in each crop. Were there any differences in Net GWP between years?

A: Thank you for your comment. The SOCSR was calculated through the long-term experiment of annual average carbon sequestration rate, not on the crop seasonal scale. We added a new Table 4 for understanding the Eo and Ei components. The GWP between years was similar.

Conclusions: you should indicate some results about wheat, not only about rice.

A: Thank you for your comment. The results include rice-wheat rotation system, not only about rice.

Lines 382-384: I guess that you are referring to one ISSM strategy in particular (N2), please indicate. Since you propose N2 as the most appropriate management strategy, I recommend describe briefly this scenario (10% reduction of N input, no rapeseed manure, no Zn or silicon addition, higher plant density...).

A: Thank you for your comment. Revised accordingly Page 16, Lines 428-429.

Table 2: please indicate with lowercase letters if there were significant differences between treatments in 2011, 2012 and 2013 (not only in the average).

A: Thank you for your comment. Revised accordingly Table 2.

Table 4: you must add the values for both wheat and rice in each component, and discuss in the text. Additionally, see my comment on lines 343-344.

A: Thank you for your comment. We added a new Table 4 for understanding the Eo and Ei components. We use the replicates for understand the uncertainties. There must have other uncertainties in selecting parameters. This is only preliminary evaluation within our understanding. Thank you very much for your great support!

Figure 2: You should delete figure 2a (this information is given on Table 2) and c (see the comment on lines 286-287). Add the standard errors (or deviations) to the columns in fig. 2b.

A: Thank you for your sincere advice! Revised accordingly Figure 1.

Figures 3 and 4. Indicate (e.g. with arrows) the times of N fertilization and the flooding cycles. A: Thank you for your comment. Revised accordingly Figures 2 and 3.

TECHNICAL CORRECTIONS

Abstract: A widespread criteria is that non-standard or uncommon abbreviations should be avoided in the abstract. Therefore, you should remove some abbreviations that you only use once, e.g. Ei, Eo and SOC.

A: Thank you for your comment. Revised accordingly Page 2, Lines 22-23.

Avoid abbreviations at the beginning of the sentence (e.g. K in line 102, CO2 in line 206, CH4 in line 263).

A: Thank you for your comment. Revised accordingly.

Line 214: The word "Conversely" does not make sense there. Please rewrite the whole sentence. A: Revised accordingly Page 10, Line 245-246.

Line 224: at the beginning of the sentence, change "A" by "The".

Line 312: Although there were not significant differences... A: Revised accordingly Page 13, Line 352. Lines 314-316: In spite of producing. Change "considerable" by "similar". A: Revised accordingly Page 13, Line 355. Line 333: per unit OF grain produced.

A: Revised accordingly Page 14, Line 375.

A: Revised accordingly Page 10, Line 255.

Line 355: the word "technologies" is repeated twice.

A: Revised accordingly Page 15, Line 399.

Line 362: the word "increment" is repeated twice.

A: Revised accordingly Page 15, Line 404.

Caption Table 2: and yields during rice and wheat cropping seasons...

A: Revised accordingly Table 2.

Footnote Table 2: significant differenceS.

A: Revised accordingly Table 2.

Table 3: I guess that you have considered MEAN fluxes and yields during the three cropping seasons (please indicate in the caption).

A: Yes, we considered mean fluxes and yields during the three cropping seasons. Revised accordingly Table 3.

Caption Table 4: global warming potential (without "s")

A: Revised accordingly Table 5.

Be careful with the font and size in the figure captions.

A: Yes, we greatly revised the whole manuscript according to your valuable comments. We also improved our Figures for clarity. Thank you very much for your great help!

Thank you once again for your critical comments and great support! Wish you a Happy New Year!

Sincerely yours, Zhengqin (on behalf of all authors)

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1	Global warming potential and greenhouse gas intensity in rice agriculture driven by		删除的内容: Net g
2	high yields and nitrogen use efficiency: A 5-year field study		
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4	Xiaoxu Zhang ^a , Xin Xu ^a , Yinglie Liu ^a , Jinyang Wang ^{a,b} , Zhengqin Xiong ^{a,*}		删除的内容: Ziqiang Zhou
5	^a Jiangsu Key Laboratory of Low Carbon Agriculture and GHGs Mitigation, College of	\searrow	删除的内容: Xin Xu ^a ,
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19	Abstract: Our understanding of how global warming potential (GWP) and greenhouse gas
20	intensity (GHGI) is affected by management practices aimed at food security with respect to
21	rice agriculture remains limited. In the present study, a 5-year field experiment was conducted
22	in China to evaluate the effects of integrated soil-crop system management (ISSM) mainly
23	consisting of different nitrogen (N) fertilization rates and split, manure, Zn and Na2SiO3
24	fertilization and planting density on GWP and GHGI after accounting for carbon dioxide (CO ₂)
25	<u>equivalent</u> emissions from all sources <u>including</u> methane, (CH ₄) and nitrous oxide, (N ₂ O)
26	emissions, agrochemical inputs, and farm operations, and sinks (i.e., soil organic carbon,
27	sequestration). For the improvement of rice yield and agronomic nitrogen use efficiency (NUE),
28	four ISSM scenarios consisting of different N rates relative to the local farmers' practice (FP)
29	rate were carried out, namely, N1 (25% reduction), N2 (10% reduction), N3 (FP rate) and N4
30	(25% increase). The results showed that compared with the FP, the four ISSM scenarios
31	significantly increased the rice yields by 10, 16, 28 and 41% and the agronomic NUE by 75, 67,
32	74 and 73%, respectively. In addition, compared with the FP, the N1 and N2 scenarios
33	significantly reduced the GHGI by 14 and 18%, respectively, despite similar GWPs. The N3
34	and N4 scenarios remarkably increased the <u>GWP</u> and GHGI by an average of 67 and 37%,
35	respectively. In conclusion, the ISSM strategies are promising for both food security and
36	environmental protection, and the ISSM scenario of N2 is the optimal strategy to realize high
37	yields and high NUE together with low environmental impacts for this agricultural rice field.
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61 1 Introduction

Rapid population growth and economic development place a growing pressure on increasing 62 food production (Barrett, 2010). An increase in global food production of 100% is the most 63 appropriate way to sustain the increase in human population and the consumption of animal 64 protein (Tilman et al., 2011). Rice is the staple food for nearly 50% of the world's people, 65 66 mainly in Asia. According to FAO (2010), approximately 600 million people in Asia-Pacific 67 region are suffering from hunger and malnutrition. With the region's population projected to 68 increase by another billion by mid-century, new approaches to increase food production are 69 needed. Within a limited land area, the intensive agricultural regions of China are facing serious environmental problems due to large inputs of chemical fertilizers and low nitrogen 70 use efficiency (NUE) (Ju et al., 2009; Makino, 2011). Thus, integrated soil-crop system 71 management (ISSM), which redesigns the whole production system based on the local 72 73 environment and draws on appropriate fertilizer varieties and application ratios, crop densities and advanced water regime management, has been advocated and developed to 74 75 simultaneously increase crop productivity and NUE with low carbon dioxide (CO₂) 76 equivalent emissions in China (Chen et al., 2014). 77 CO_2 methane (CH₄) and nitrous oxide (N₂O) are the most important greenhouse gases (GHGs) that greatly contribute to global warming (IPCC, 2013). The concept of global 78 79 warming potential (GWP) was proposed based on the radiative properties of all the GHG 80 emissions and soil organic carbon (SOC) fixation, expressed as CO₂ eq. ha⁻¹ yr⁻¹ (Robertson and Grace, 2004; Mosier et al., 2006). Although agriculture releases significant amounts of 81 82 CH₄ and N₂O into the atmosphere, the net emission of CO₂ equivalents from farming 83 activities can be partly offset by changing agricultural management to increase the soil organic matter content and/or decrease the emissions of CH4 and N2O (Mosier et al., 2006; 84 Smith et al., 2008). If global agricultural techniques are improved, the mitigation potential of 85 agriculture (excluding fossil fuel offsets from biomass) is estimated to be approximately 86

5.5–6.0 Pg CO₂ eq. yr⁻¹ by 2030 (Smith et al., 2008). However, the release of CO₂ 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). This indicates that agricultural ecosystems are not only a very important source of GHG emissions 删除的内容: carbon (C) costs 删除的内容: Carbon dioxide (删除的内容:) but also present substantial opportunities for mitigation. Therefore, when determining the
<u>GWP, of GHG (CO₂, CH₄ and N₂O) emissions from agroecosystems, there is a need to
account for all sources <u>including_GHGs</u> emissions, agrochemical inputs (Ei) and farm
operations (Eo) and sinks_e.g, soil organic carbon (SOC) sequestration of <u>CO₂</u> equivalents
(Sainju et al., 2014).
</u>

99 Information on the effects of ISSM scenarios on GWP and greenhouse gas intensity 100 (GHGI) is limited in China (Ma et al., 2013; Liu et al., 2015). The annual rotation of summer 101 rice-upland crop is a dominant cropping system in China. Previous studies mainly 102 investigated the initial influences of ISSM practices but did not account for the contributions of CO₂ emissions from Ei and Eo (Ma et al., 2013; Zhang et al., 2014). In this study, we 103 104 evaluated <u>GWP</u> and GHGI by taking CO₂ equivalents from all sources and sinks into account for 5 years. We hypothesized that the ISSM strategies would reduce the overall GWP and 105 106 GHGI compared with local farmers' practices (FP), The specific objectives of this study were to (i) evaluate the effects of different ISSM scenarios on <u>GWP</u> and GHGI; (ii) determine the 107 main sources of GWP and GHGI in a rice-wheat cropping system; and (iii) elucidate the 108 overall performance for each ISSM scenario for different targets to increase grain yields and 109 110 NUE and reduce <u>GWP</u> and GHGI.

111 2 Materials and Methods

112 2.1 Experimental site

113 A 5-year field experiment was conducted at the Changshu agro-ecological experimental 114 station (31°32'93"N, 120°41'88"E) in Jiangsu Province, China. This is a typical, intensively 115 managed agricultural area where the cropping regime is dominated by a flooding rice (Oryza 116 sativa L.)-drained wheat (Triticum aestivum L.) rotation system. The site is characterized by a 117 subtropical humid monsoon climate with a mean annual air temperature of 15.6, 15.2 and 15.8, °C and precipitation of <u>878, 1163 and 984 mm for three years, respectively</u>. The soil of 118 119 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. 120 121 The major properties of the soil at 0-20 cm can be described as follows: bulk density, 1.11 g

122 cm^{-3} ; pH, 7.35; organic matter content, 35.0 g kg⁻¹; and total N, 2.1 g kg⁻¹. The daily mean air

temperatures and precipitation during the study period from June 15, 2011, to June 15, 2014,

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scenarios can be adopted by policy makers
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147	are given in the supplementary resource 1,	删除的内律
148	2.2 Experimental design and management	
149	A completely randomized design was established in 2009 with four replicates of six	
150	treatments, including no nitrogen (NN) and FP as controls, and four ISSM scenarios at	
151	different N application rates relative to the local FP rate, namely N1 (25% reduction), N2 (10%	
152	reduction), N3 (FP rate) and N4 (25% increase). The designed ISSM (only for rice but not	
153	wheat production) including a redesign of a split N fertilizer application, a balanced fertilizer	
154	application (rapeseed cake in additional 112.5 kg N ha ⁻¹ , C/N=8), additional phosphorus and	
155	potassium application, and transplanting density, used as the main techniques for improving	
156	rice yield and agronomic NUE (calculated as the difference in grain yield between the plots	
157	that received N application and the NN plot, divided by the N fertilizer rate). The details of	
158	the fertilizer applications, irrigation, and field management practices of the six different	
159	treatments are presented in Table 1. Further detailed information was described previously	
160	(Zhang et al., 2014). Each plot was 6 m \times 7 min size with an independent drainage/irrigation	
161	system.	
162	One midseason drainage (about one week) and final drainage before harvest were used	
163	during the rice-growing season, whereas the plots only received precipitation during the	
164	wheat-growing season. The N fertilizer was split into a 6:2:0:2 or 5:1:2:2 ratio of basal	
165	fertilizer and topdressings for the rice crop and a 6:1:3 ratio for the wheat crop. All of	
166	phosphorous (P), silicon (Si), zinc (Zn), and rapeseed cake manure were applied as basal	删除的内容
167	fertilizers for both crops. <u>Potassium (K)</u> was added as a split (1:1) application to the rice crop	
168	and <u>all</u> as basal fertilizer for the wheat crop. The basal fertilization occurred at the time of rice	删除的内容
169	transplanting and wheat seeding. The topdressing was applied at the tillering and panicle	
170	stages of the rice crop and at the seedling establishment and elongation stages of the wheat	
171	crop. Harvests included crop grains as well as the rice and wheat straws were removed out of	
172	the field for all the treatments in this study.	
173	2.3 Gas sampling and measurements	
174	We measured the CH_4 emissions and N_2O fluxes in each plot of the field experiment over five	
175	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 176 12

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in our previous study (Ma et al., 2013). Emissions were measured manually using the 180 删除的内容: a hydrogen flame ionization detector and an SS-2 m×2 mm Porapak Q static-opaque chamber method. Each replicate plot was equipped with a chamber with a size 181 (80/100 mesh) column. The oven of 50 cm \times 50 cm \times 50 cm or 50 cm \times 50 cm \times 110 cm, depending on the crop growth and 182 temperature remained at 50 °C, and the detector was maintained at 300 °C. The 183 plant height. The chamber was placed on a fixed PVC frame in each plot and wrapped with a carrier gas was purified N2 with a flow rate layer of sponge and aluminum foil to minimize air temperature changes inside the chamber 184 of 35 ml min⁻¹ 185 during the period of sampling. 删除的内容: NGWP 186 The gas samples were analyzed for CH₄ and N₂O concentrations using a gas 删除的内容: In addition to the CH emissions and N2O fluxes, we considered 187 chromatograph (Agilent 7890A, Shanghai, China) equipped with two detectors. CH₄ was some 'hidden' CO2 equivalent emissions. 188 detected using a hydrogen flame ionization detector (FID), and N2O was detected using an including agrochemical inputs (Ei), such as the manufacture and transportation of the N. electron capture detector (ECD). Argon-methane (5%) and N2 were used as the carrier gas at a 189 P and K fertilizers (Snyder et al., 2009), and flow rate of 40 ml min⁻¹ for N₂O and CH₄ analysis, respectively. The temperatures for the farm operations (Eo), such as the water 190 used for irrigation (Zhang et al., 2013) and column and ECD detector were maintained at 40 °C and 300 °C, respectively. The oven and 191 diesel fuel (Huang et al., 2013a). The CO₂ 192 FID were operated at 50 °C and 300 °C, respectively, equivalent emissions of N fertilizer were calculated as the mean value of the C 193 2.4 Topsoil organic carbon sequestration measurements emissions of 1.3 kg C equivalent kg⁻¹ (Lal, To measure the organic carbon content of the topsoil as described by Zhang et al. (2014), soil 194 2004). Similarly, the CO₂ equivalent for irrigation was calculated from the total samples were collected after the wheat harvest in 2009 and 2014 from all experimental plots 195 amount of water used during the 196 at a plowing depth of 0-20 cm. The soil organic carbon sequestration rates (SOCSR) were rice-growing season; the coefficient for the C cost was 5.16 (kg C eq. $cm^{^{-1}} ha^{^{-1}})$ (Lal, 197 calculated as follows (Liu et al., 2015): 2004). The CO2 equivalents of other Ei (P SOCSR (t C ha⁻¹ yr⁻¹) = (SOC_t - SOC₀) / T × γ × (1 - δ_{2mm} /100) × 20 × 10⁻¹ (1) 198 and K fertilization, manure, herbicide, pesticide and fungicide applications) and In Eq. (1), SOC_t and SOC₀ are the SOC contents measured in the soils sampled after the 199 Eo (tillage, planting, harvest, and farm 200 wheat was harvested in 2014 and 2009, respectively. T refers to the experimental period (yr). machinery production) were recorded and estimated according to the methods 201 γ and δ_{2mm} are the average bulk density and the gravel content (>2 mm) of the topsoil (0–20 删除的内容: The NGWP of the cropland 202 cm), respectively. ecosystem equals the total CO2 emission equivalents minus the SOC change. To 203 2.5 GWP and GHGI measurements better understand climatic effects of the To better understand the overall climatic effects of the ISSM strategies on rice-wheat 204 删除的内容: NGWP rotation cropping system, the GWP and GHGI were updated using all possible components 205 删除的内容: using and calculated as the following equations (IPCC, 2013): 206 删除的内容: GWP(<u>GWP</u> (kg CO₂ eq. ha⁻¹ yr⁻¹) = $25 \times CH_4 + 298 \times N_2O + Ei + Eo - 44/12 \times SOCSR_(2)$ 207 删除的内容: GWP(GHGI (kg CO₂ eq. kg⁻¹ grain yield yr⁻¹) = <u>GWP</u>/grain yield (3) 208 删除的内容:) In Eq. (2), Ei, Eo and SOCSR represent CO_2 equivalent emissions from the 删除的内容: NGWP 209

281	agrochemical inputs, farm operations and soil organic carbon sequestration rate, respectively.
282	The global warming potential of 1 kg CH_4 and N_2O are equivalent to 25 and 298 kg CO_2
283	based on 100-year time scale, respectively (IPCC, 2013). The 12 and 44 are the molecular
284	weight of C and CO_2 , respectively. The grain yield is expressed as the air-dried grain yield.
285	Therefore, the GWP of the cropland ecosystem equals the total CO ₂ equivalent emissions
286	minus the SOC change. In addition to the CH ₄ emissions and N ₂ O fluxes, we considered the
287	'hidden' CO2 equivalent emissions, including agrochemical inputs (Ei), such as the
288	manufacture and transportation of the N, P and K fertilizers (Snyder et al., 2009), and farm
289	operations (Eo), such as the water used for irrigation (Zhang et al., 2013) and diesel fuel
290	(Huang et al., 2013a). The CO2 equivalent emissions of N fertilizer were calculated as the
291	mean value of the C emissions of 1.3 kg C equivalent kg ⁻¹ (Lal, 2004). Similarly, the CO_2
292	equivalent for irrigation was calculated from the total amount of water used during the
293	rice-growing season; the coefficient for the C cost was 5.16 (kg C eq. $cm^{-1} ha^{-1}$) (Lal, 2004).
294	The CO ₂ equivalents of other Ei (P and K fertilization, manure, herbicide, pesticide and
295	fungicide applications) and Eo (tillage, planting, harvest, and farm machinery production)
296	were recorded and estimated according to the methods provided by Lal (2004). We collected
297	data specific to China's fertilizer manufacture and consumption, and then estimated C
298	emissions coefficients were 0.07 and 0.1 C cost (kg C eq. kg ⁻¹ active ingredient) per applied
299	Si and Zn fertilizer, respectively.
300	2.6 Statistical analysis
301	Repeated-measures multivariate analysis of variance (MANOVA) and linear relationships
302	were determined using JMP 7.0, ver. 7.0 (SAS Institute, USA, 2007). The F-test was applied
303	to determine whether there were significant effects of the practices, years and their interaction
304	at $P < 0.05$. One-way analysis of variance was conducted to determine the emissions of CH ₄
305	and N2O, and the grain yield among the different treatments. Tukey's HSD test was used to
306	determine whether significant differences occurred between the treatments at a significance
307	level of $P < 0.05$. The results are presented as the means and standard deviation (mean ±SD,
308	n = 4).
309	3 Results

310 3.1 Crop production and agronomic NUE

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删除的内容: using JMP, ver. 7.0 (SAS Institute, USA, 2007)

删除的内容: Repeated-measures multivariate analysis of variance (MANOVA)Two-way analysis of variance (ANOVA) and linear relationships were determined using JMP 7.0. The F-test was applied to determine whether there were significant effects of the practices, years and their interaction at P < 0.05.</p> 321 During the three cropping rotations from 2011 to 2014, the rice and wheat yields varied significantly among these cultivation patterns; these results are shown in Table 2. The grain 322 yields ranged from 5.83 to 12.11 t ha^{-1} for rice and 1.75 to 6.14 t ha^{-1} for wheat (Table 2). On 323 324 average over the three cycles, the annual rice yield of the FP was significantly lower than that of the ISSM scenarios of N1, N2, N3 and N4. Compared with the FP, rice grain yields 325 326 increased by 10% and 16% for the N1 and N2 scenarios, respectively, i.e., with the lower N 327 input, by 28% for the N3 scenario with the same N input and by 41% for the N4 scenario with 328 the highest N input. However, we did not observe any significant increases in the wheat-grain 329 yields compared with the FP except for the N4 scenario. Statistical analysis indicated that rice 330 and wheat yields from the three years were not significantly influenced by the interaction of

- 331 <u>cultivation patterns and cropping year (Table 3).</u>
- The agronomic NUE for the rice and wheat of the fertilized plots ranged from 9.2 to 16,1and 19.5 to 24.7 kg grain kg⁻¹ N, respectively (Fig. <u>1</u>). The higher NUE in the wheat season was mainly due to the reduced N fertilizer (40%) during this season. As expected, the rice agronomic NUE significantly increased by 75, 67, <u>74</u>, and <u>73%</u> for the N1, N2, N3 and N4 scenarios, respectively, compared with the FP (Fig. <u>1</u>). For the wheat crop, the agronomic NUE merely increased by 12 and 14% for the N1 and N2 scenarios, respectively, and decreased to some extent for the N3 and N4 scenarios compared with the FP, mainly because
- 339 the current ISSM strategy was only designed for rice and not wheat production.
- 340 3.2 CH₄ and N₂O emissions
- 341 All plots showed similar CH_4 emission patterns, being a source in the rice season and
- 342 negligible in the wheat season. During the three annual rice-wheat rotations from 2011 to
- 343 2014, the CH₄ fluxes ranged from -3.89 to 99.67 mg C m⁻² h⁻¹ (Fig. 2). The seasonal CH₄
- 344 emissions varied significantly among the treatments during the rice-growing season (Table 3,

Fig. <u>2</u>). No significant difference was found between the FP, N1 and N2 plots. <u>Temporal</u>
<u>variation was significant during the three cycles (Table 3, P < 0.001).</u> Averaged across years,

- 347 the CH_4 emission was greater in the N3 and N4 plots than in the NN, FP, N1 and N2 plots
- 348 (Table 2, P < 0.05). However, compared with the NN plots, the FP, N1 and N2 plots with
- 349 inorganic fertilizer application resulted in increased CH_4 emission rates of 59.9, 41.9 and
- 43.0%, respectively, averaged over the rice-growing seasons. The CH_4 emission rates were

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	ther enhanced by 198.5% in the N3 plots and by 246.7% in the N4 plots	359
	The annual N2O fluxes varied from –33.1 to 647.5 μg N2O-N m^-2 $h^{-1}{}_{a}$ most of the N2O	360
C	s emitted during the wheat-growing season after fertilization events, and there were several	361
	all emission peaks during the rice-growing season (Fig. 3). With respect to the N	362
	lication effect, the annual cumulative N2O emissions for all four ISSM scenarios were	363
	nificantly higher than in NN ($P < 0.05$). Relative to the FP plot, the N1 and N2 scenarios	364
	reased the annual N ₂ O emissions by an average of 41% and 22%, respectively <u>, (Table 2).</u>	365
	e N4 scenario significantly increased it by 46% ($P < 0.05$) because they received additional	366
	via manure application compared to the FP practice, although there was no significant	367
	Ference between the N3 and FP plots.	368
	Annual GWP and GHGI	369
	sed on the perspective of the carbon footprint, we included the GHG emissions associated	370
	h all of the inputs (Ei and Eo), and SOC sequestration was expressed as kg CO_2 eq. ha^{-1}	371
	. The emission of CO_2 equivalents for Ei and Eo are classified in Table 4. While irrigation	372
	s a large proportion of farm operations, these were much less significant than chemical	373
	uts. The CO_2 equivalents rates from N fertilizer dominated not only the chemical input	374
	tion ($67_{\overline{r}}$ -76% of Ei) but also the total CO ₂ equivalents from agricultural management	375
\langle	-51% of the sum of the Ei and Eo). The <u>GWP</u> ranged from 7871 to 20911 , kg CO ₂ eq. ha ⁻¹	376
	for the NN and the N4 plots, respectively (Table 5). Although fertilized treatments,	377
	reased the annual CH_4 and N_2O emissions, it also increased the SOC sequestration in this	378
	pping system. Of the main field GHGs that were directly emitted, CH_4 accounted for	379
//}	-75% of the <u>GWP</u> in all plots. An increase in the annual SOC content led to a significant	380
// }	rease in the <u>GWP</u> (contributed to 5–10% of the <u>GWP</u> except in the NN plot). The CO ₂ $/$	381
\square	vivalents from agricultural management practices, for Ei (2449-4256, CO ₂ eq. ha ⁻¹ yr ⁻¹)	382
λ	re higher than those for Eo (1285–1697CO ₂ eq. ha^{-1} yr ⁻¹) in the fertilized plots. There was	383
	significant difference in the annual <u>GWP</u> observed between the FP, N1 and N2 plots (Table	384
	Across the three years, N1 and N2 slightly reduced the <u>GWP</u> by 12 and 10%, respectively; $/$	385
	wever, N3 and N4 significantly increased the GWP by an average of 52 and 81%,	386
	pectively, in comparison with the FP.	387
	The GHGI was used to express the relationship between GWP and grain yield. The	388

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NGWPGWP was achieved under the N1
scenario for the ISSM.
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The GHGI was used to express the relationship between <u>GWP</u> and grain yield. The 16

418	GHGIs in this study ranged from 664 to 1145 , kg CO ₂ eq. t ⁻¹ (Table 5). The significant
419	difference in the annual GHGI was found between the FP and the ISSM strategies. Compared
420	with the FP, N1 and N2 significantly reduced the GHGI by 14 and 18%, respectively, mainly
421	due to the increased grain yield and SOC sequestration as well as reduced GHG emissions for
422	the ISSM strategies. Although N fertilizer or organic/inorganic combination fertilizer
423	application reduced the SOC losses caused by crop cultivation and increased the grain yields,
424	the GHGIs were generally higher for the N3 and N4 scenarios than the N1 and N2 scenarios
425	due to further increases in CH_4 and N_2O emissions.
426	4 Discussion
427	4.1 Grain yield and agronomic NUE as affected by ISSM strategies
427 428	4.1 Grain yield and agronomic NUE as affected by ISSM strategies Grain yields are directly related to fertilizer management. The <u>MANOVA results indicated</u>
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428	Grain yields are directly related to fertilizer management. The MANOVA results indicated
428 429	Grain yields are directly related to fertilizer management. The <u>MANOVA results indicated</u> that the rice and wheat grain yields were significantly affected by the cultivation strategies
428 429 430	Grain yields are directly related to fertilizer management. The <u>MANOVA results indicated</u> 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
428 429 430 431	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 plot, the rice yields were remarkably increased by all four

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First, the adjusted transplanting density for the N1, N2, and N3 scenarios would produce a

positive effect on rice yield by influencing rice colony structure, which agreed with Wu et al. (2005). Second, reasonable N split for the N1, N2, N3 and N4 scenarios would significantly

increase rice yield and agronomic NUE which had been confirmed by Liu et al. (2009). In the

present study, N1 and N2 significantly increased annual rice production by 10 and 16%, respectively, in comparison with the FP (Table 2). The finding is consistent with the result 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 N3 and N4 scenarios

would support better seedling establishment and reduce both biotic and abiotic stress, thus

produce 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
increased by 25% and applied with rapeseed cake manure, the rice yield in these N3 and N4
plots remarkably increased 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, which improved the rice yield, v

457 It has been suggested that N losses vary depending on the timing, rate, and method of N 458 application, as well as the source of N fertilizer (Zhu, 1997). In spite of the high proportion and improper timing of N application, rapid N losses (via ammonia volatilization, 459 denitrification, surface runoff, and leaching) are important factors that cause low agronomic 460 NUE of irrigated rice in China (Peng et al., 2006). Compared with the FP plot, the rice 461 agronomic NUE was significantly increased by 75, 67, 74 and 73% under the N1, N2, N3 and 462 N4 scenarios, respectively (Fig. 1). The higher rice agronomic NUE in our study over the 463 experimental period was primarily due to the greatly reduced N losses by leaching and 464 465 volatilization as well as the improvement of N bioavailability in the rice crop season. 466 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 467 strategy is an effective method for improving grain yield and agronomic NUE for future 468 sustainable rice agriculture in China. 469

470 4.2 CH₄ and N₂O emissions as affected by ISSM strategies

During the three years, the annual cumulative CH₄ emissions, on average, varied from 133 to 471 469 kg C ha⁻¹yr⁻¹ (Table 2), and these values fell within the range of 4.1 to 1015.6 kg CH₄ 472 473 ha⁻¹ observed previously in a rice field (Huang et al., 2004). The MANOVA results indicated 474 that obvious effects of cultivation patterns<u>and years</u> on CH₄ emissions were found during the 475 rice-wheat rotations (Table 3, P < 0.001). The CH₄ emissions were not significantly affected by the cycles but affected by crop season (Table 5, Fig. 2). In this study, no significant 476 477 difference in CH₄ emission was observed between the FP, N1 and N2 plots. However, 478 compared with the FP plot, the N3 and N4 scenarios emitted 87 and 118% more CH₄ emissions, respectively (Table 5), which is probably due to the incorporation of the organic 479 18

删除的内容: and the efficient control of pests and diseases (Slaton et al., 2005; Wang et al., 2005)

删除的内容: The higher yields in N3 and N4 scenarios would have also attributed to the addition of 15 kg Zn ha⁻¹ for better seedling establishment and vigor according to Slaton et al. (2005) and the application of silicon to reduce both biotic and abiotic stress according to Wang et al. (2005).

删除的内容: In general, rapid N losses (via ammonia volatilization, denitrification, surface runoff, and leaching), the high application rate of fertilizer N and improper timing of N application are three important factors that cause low agronomic NUE of irrigated rice in China

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502	rapeseed cake manure. Previous reports support the observations that CH_4 emissions were
503	significantly increased with the application of organic amendments (Ma et al., 2009;
504	Thangarajan et al., 2013; Zou et al., 2005). Additional application of Si and Zn fertilizers had
505	no significant effect on CH_4 and N_2O fluxes, which was consistent with the result of Xie et al.
506	(2015). Moreover, rice growth was found to be significantly increased under the N3 and N4
507	scenarios. In this case, the organic matter inputs such as root litter and rhizodeposits in the N3
508	and N4 scenarios were probably also higher than in the other plots, and thus soil C input,
509	which served as an additional source of substrates for the methanogens in the rice paddies,
510	likely contributed to the increase in CH ₄ emissions (Ma et al., 2009). Finally, because the rice
511	plants acted as the main pathway for CH4 transports from the soil to the atmosphere, the
512	higher biomass the more CH ₄ emissions (Yan et al., 2005). The results obtained in the present
513	study revealed that both inorganic and organic fertilizer application significantly increased the
514	CH ₄ emissions in the rice season (Table 2), which was probably associated with the increase
515	in the SOC content and crop biomass (Ma et al., 2013).
516	Denitrification and nitrification are the main processes that produce N ₂ O in the soil (Paul

et al., 1993). Changes in the soil water content strongly affected the soil N2O emissions and 517 518 resulted in negligible N2O emissions when the rice field was flooded (Fig. 3), which is consistent with previous reports (Akiyama et al., 2005; Murdiyarso et al., 2010). A relatively 519 520 high N₂O peak was observed in the first two weeks of the wheat-growing season (Fig. 3), 521 possibly because soil changes from flooded to drained condition may have enhanced N2O 522 release (Deng et al., 2012). Alternation of drainage and flooding may induce large amounts of 523 N₂O emissions, particularly in fertilized systems; this has commonly been proved in earlier 524 studies (Wang et al., 2013; Xiong et al., 2007; Zou et al., 2005). The seasonal and annual rates 525 of N2O emission were significantly affected by the cultivation practice patterns and years (Table 3). Compared with the FP plot, the N2 scenario greatly decreased the seasonal N2O 526 527 emissions in this study, which may have resulted from a reduction in the N fertilizer rate (Table 1, Table 2). The total N_2O emissions decreased by 7–38% and 26–42% in the rice and 528 wheat seasons, respectively, when the conventional N management (300 kg N ha⁻¹ for rice 529 and 180 kg N ha⁻¹ per crop for wheat) changed to optimum N management (225-270 kg N 530 ha^{-1} for rice and 135–162 kg N ha^{-1} per crop for wheat). It is likely that more N₂O was 531 19

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emitted (Mosier et al., 2006) as a result of the additional N made available to the soil
microbes through N fertilizer application, which probably increased the CH₄ emissions
(Banger et al., 2013). Strategies that can reduce N fertilization rates without influencing crop
yields can inevitably lower GHG emissions (Mosier et al., 2006).

542 4.3 <u>GWP</u> and GHGI as affected by ISSM strategies

The <u>GWP</u> in our study (10104–20911, kg CO_2 eq. ha⁻¹) with the ISSM strategies was higher 543 544 than that in a double-cropping cereal rotation (1346–4684 kg CO_2 eq. ha⁻¹) and a rice-wheat 545 annual rotation (290–4580 kg CO_2 eq. ha⁻¹) reported by Huang et al. (2013b) and Yang et al. 546 (2015), respectively. Dominant CH₄ emissions as well as additional CO₂ emitted by the 547 machinery/equipment used for irrigation and farm operations under the ISSM strategies may 548 increase the <u>GWP</u> more than in other cropping systems. However, the current <u>GWP</u> was still much lower than that of a double-rice cropping system (13407–26066 kg CO_2 eq. ha⁻¹) 549 (Shang et al., 2011). The GHGIs, which ranged from 0.66 to 1.15 kg CO₂ eq. kg⁻¹ grain in this 550 study, were slightly higher than previous estimates of 0.24-0.74 kg CO₂ eq. kg⁻¹ grain from 551 552 rice paddies with midseason drainage and organic manure incorporation (Qin et al., 2010; Li 553 et al., 2006) but were lower than the DNDC model estimates for continuous waterlogged paddies (3.22 kg CO_2 eq. kg⁻¹ grain) (Li et al., 2006). Differences in <u>GWP</u> or GHGI were 554 found in the cultivation patterns over the three rice-wheat rotations (Table 5). Although there 555 were not significant differences among the FP, N1 and N2 plots, the N1 and N2 scenarios with 556 557 optimized ISSM strategies led to a lower GWP than the FP (Table 5). Compared with the FP, 558 the N1 and N2 scenarios dramatically reduced the GHGI, which was mainly due to higher 559 vields. In spite of the similar, GWP compared with the FP plot, the lowest GHGI (0.66 kg CO₂ eq. kg⁻¹ grain) was obtained under the N2 scenario. This finding is consistent with the 560 suggestion made by Burney et al. (2010), i.e., that the net effect of higher yields offsets 561 emissions. It is well known that CH₄ emissions dominate the <u>GWP</u> in rice paddies (Ma et al., 562 2013; Shang et al., 2011). In comparison to the <u>GWP</u> (11545 kg CO₂ eq. ha⁻¹yr⁻¹) and GHGI 563 (0.81 kg CO_2 eq. kg⁻¹ grain) of the FP, the N3 and N4 scenarios increased both the GWP and 564 GHGI, mainly because these scenarios notably increased the CH₄ emissions compared with 565 the FP, which resulted in relatively higher <u>GWP</u> (Table 5). 566

567 Agricultural management practices that change one type of GWP source/sink may also 20

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583	impact other sources/sinks and therefore change the GWP and GHGI (Mosier et al., 2006;	\square	删除的内容: NGWP
584	Shang et al., 2011). Although the N-fertilizer plots, especially those with the incorporation of		
585	organic fertilizer, increased the annual CH_4 and N_2O emissions, they increased the SOC		
586	sequestration in this cropping system, which is agreement with previous reports (Huang and		
587	Sun, 2006). This was mainly due to the enhanced incorporation of rapeseed cake and crop		
588	residue associated with higher crop productivity (Ma et al., 2013). In the present study, the N2		
589	scenario with ISSM decreased the CH_4 and $\mathrm{N}_2\mathrm{O}$ emissions as well as the energy consumption		
590	related to irrigation and the manufacture and transport of N fertilizer (depending on coal		
591	combustion), ultimately leading to a decrease in the <u>GWP</u> relative to the FP plot. Moreover,		删除的内容: NGWP
592	despite the lower N fertilizer input, the grain yield did not decline and the GHGI of the N2		
593	scenario was thus lower than of the FP plot, indicating less consumption of CO_2 equivalents		
594	per unit \underline{of} grain produced. We demonstrate that high yield and agronomic NUE, together with		
595	low GWP, are not conflicting goals by optimizing ISSM strategies.		
596	4.4 Main components of GWP and GHGI and implementation significance for the ISSM	\square	删除的内容: NGWP
597	strategies		
598	Determining the main components of the GWP and GHGI in specific cropping systems is		删除的内容: NGWP
599	very important for mitigating GHG emissions in the future because the benefits of C		
600	sequestration would be negated by \mbox{CH}_4 and N_2O emissions and the \mbox{CO}_2 equivalents released		
601	with the use of high N fertilizer application rates (Schlesinger, 2010). In the current study, the		
602	five main components of the CO_2 equivalents for the <u>GWP</u> were ranked in decreasing order of	\square	删除的内容: NGWP
603	importance as follows: CH_4 emissions > agrochemical inputs of N fertilizer > farm operations		
604	related to irrigation > SOC sequestration > N_2O emissions (Table 5). <u>CH₄ emissions, the most</u>		
605	important component of GWP in this typical rice-wheat rotation system, could be further		
606	mitigated by some other strategies, such as reasonable irrigation (Zou et al., 2005; Wang et al.,		
607	<u>2012).</u>		
608	Although N fertilizer application increased SOC sequestration when it was applied		
609	with rapeseed cake manure, this benefit was consistently overshadowed, on a CO_2 equivalent		
610	basis, by the increases in CH_4 and N_2O emissions (Table 5). Similar results have been		
611	reported, i.e., GHG emissions substantially offset SOC increases (Six et al., 2004). It is		删除的内容: that
612	possible that the realization of reducing the <u>GWP</u> and GHGI in China should focus on		删除的内容: NGWP
	21		

increasing the SOC and simultaneously decreasing the CO_2 equivalents from CH_4 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).

627 China is a rapidly developing country that faces the dual challenge of substantially 628 increasing grain yields 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 629 rice production system that achieved mean yields of 10.63 t ha^{-1} (an increment of almost 24%) 630 and an agronomic NUE of 16.33 kg grain kg⁻¹ N (an approximate doubling) in long-term field 631 632 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 633 appropriate fertilizer varieties and application ratios, crop densities and an advanced water 634 regime management. If the ISSM strategies were also developed for the rotated wheat crop, 635 636 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 637 strategies are promising, particularly the ISSM-N2 scenario, which is the most favorable to 638 639 realize higher yields with lower environmental impact. The proposed ISSM strategies can 640 provide substantial benefits to intensive agricultural systems and can be applied feasibly using 641 current technologies.

642 5 Conclusions

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 <u>GWP</u> and GHGI affected by different ISSM scenarios. After a five-year field experiment, we found that the CH₄ emissions, production of N fertilizer, irrigation, SOC sequestration and N₂O fluxes were the main components of the <u>GWP</u> in a typical rice-wheat rotation system. In contrast with the FP, N1 and N2 significantly reduced the GHGI, though they resulted in similar <u>GWPs</u>, and N3 and N4 remarkably increased the <u>GWP</u> and GHGL By

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research is required for mitigating GHG
emissions when aiming to increasing rice

yield and NUE

659	adopting the ISSM strategy, the conventional N application rate was reduced by 10% while
660	the rice yield was significantly increased by 16%, the NUE was improved by 67% and the
661	GHGI was lowered. ISSM scenarios could be adopted for both food security and
662	environmental protection with specific targets. We propose that the ISSM-N2 scenario is the
663	most appropriate management strategy (10% reduction of N input, no rapeseed manure and
664	higher plant density) for realizing higher yields and NUE, together with some potential to
665	reduce GHGI by integrated soil-crop management. For simultaneously mitigating GHG
666	emissions, further research on integrated soil-crop system managements is required
667	particularly for mitigating CH ₄ emissions in sustainable rice agriculture.

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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 ₂ O ₅ :K ₂ O:Na ₂ SiO ₃ :ZnSO ₄ , kg ha ^{-1})	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^{-1})	0	0	0	0	2.25	2.25	
Water regime	F-D-F-M ^b	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	
		Wheat-growing season					
Chemical fertilizer application rate $(N:P_2O_5:K_2O, kg ha^{-1})$	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^{-1})	180	180	180	180	180	180	

^aNN, 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⁻¹ for the rice crop and 180 kg N ha⁻¹ for the wheat crop, namely, N1 (25% reduction), N2 (10% reduction), N3 (FP rate) and N4 (25% increase). Urea,

calcium biphosphate and potassium chloride were used as N, P and K fertilizer respectively.

^bF-D-F-M, flooding-midseason drainage-re-flooding-moist irrigation.

Seasonal CH_4 and N_2O emissions, and vields during rice and wheat cropping seasons, in the three

删除的内容: ... yields during rice and

cycles of 2011-2014.

		Rice season			Wheat season	
Treatment	CH ₄	N_2O	Yield	CH ₄	N ₂ O	Yield
	(kg C ha^{-1})	(kg N ha ⁻¹)	(t ha ⁻¹)	(kg C ha^{-1})	(kg N ha ⁻¹)	(t ha ⁻¹)
2011						
NN	153 <u>+10.8c</u>	0.03±0.05 <u>c</u>	5.85±0.08 <u>f</u>	- 0.48±0.63 <u>a</u>	0.45 ±0.09 <u>d</u>	1.74±0.18 <u>d</u>
FP	26 <u>6</u> ±25. <u>3b</u>	0.11±0.08 <u>c</u>	8.38±0.35 <u>e</u>	- 0.48±1.86 <u>a</u>	1.43±0.19 <u>b</u>	5.67 ±0.20 <u>b</u>
ISSM-N1	212 <u>+30.3bc</u>	0.08±0.03 <u>c</u>	9.27±0.26 <u>d</u>	0.78±0.97 <u>a</u>	0.65±0.11 <u>cd</u>	5.05±0.16 <u>c</u>
ISSM-N2	2 <u>20</u> ±32.5 <u>bc</u>	0.17±0.11 <u>bc</u>	9.79±0.44 <u>c</u>	2.25±2.07 <u>a</u>	0.80±0.06 <u>c</u>	5.71±0.18 <u>b</u>
ISSM-N3	518 <u>±</u> 58.9 <u>a</u>	0.38±0.15 <u>ab</u>	10.81±0.26 <u>b</u>	0.04±3.23 <u>a</u>	1.40±0.10 <u>b</u>	5.31 ±0.26 <u>bc</u>
ISSM-N4	56 <u>1,±</u> 50. <u>9a</u>	0.37±0.07 <u>a</u>	11.76±0.24 <u>a</u>	- 0.09±1.40 <u>a</u>	1.93±0.09 <u>a</u>	6.15±0.15 <u>a</u>
2012						
NN	149 <u>+</u> 25. <u>8d</u>	0.13±0.10 <u>c</u>	5.80±0.22 <u>f</u>	- 4.32±7.29 <u>a</u>	0.65±0.09 <u>d</u>	1.73±0.11 <u>c</u>
FP	239 <u>+</u> 34.5 <u>c</u>	0.33±0.11 <u>bc</u>	8.72±0.62 <u>e</u>	4.85±10.30 <u>a</u>	2.13±0.43 <u>ab</u>	5.64±0.34 <u>ab</u>
ISSM-N1	22 <u>6</u> ±30.4 <u>cd</u>	0.27±0.07 <u>bc</u>	9.43±0.34 <u>d</u>	1.46±6.38 <u>a</u>	1.39±0.14 <u>c</u>	4.94 ±0.38 <u>b</u>
ISSM-N2	228±32.6cd	0.38±0.29 <u>bc</u>	9.99±0.50 <u>c</u>	- 1.02±0.84 <u>a</u>	1.77±0.38 <u>bc</u>	5.78±0.59 <u>ab</u>
ISSM-N3	431 <u>+26.8b</u>	0.52±0.16 <u>ab</u>	10.92±0.61 <u>b</u>	2.45±8.35 <u>a</u>	2.19±0.24 <u>ab</u>	5.39±0.39 <u>ab</u>
ISSM-N4	53 <u>6±58.7a</u>	0.78±0.13 <u>a</u>	12.24±0.60 <u>a</u>	5.91±6.18 <u>a</u>	2.61±0.42 <u>a</u>	6.10±0.49 <u>a</u>
2013						
NN	101 <u>+39.2</u> <u>h</u>	0.16±0.09 <u>b</u>	5.84±0.15 <u>f</u>	- 1.45±1.34 <u>a</u>	0.35±0.06 <u>c</u>	1.80±0.03 <u>c</u>
FP	14 <u>1+25.2</u> h	0.43±0.39 <u>ab</u>	8.67±0.26 <u>e</u>	- 3.70±1.76 <u>a</u>	0.80±0.20 <u>ab</u>	5.70±0.30 <u>ab</u>
ISSM-N1	135 <u>+15.7b</u>	0.19±0.16 <u>ab</u>	9.66±0.29 <u>d</u>	- 1.00±1.61 <u>a</u>	0.49±0.16 <u>bc</u>	5.15±0.20 <u>b</u>
ISSM-N2	129 <u>+</u> 32.2 <u>h</u>	0.26±0.13 <u>ab</u>	10.15±0.07 <u>c</u>	- 0.79±1.60 <u>a</u>	0.69±0.24 <u>abc</u>	5.80±0.18 <u>ab</u>
ISSM-N3	256 <u>±</u> 45.6 <u>a</u>	0.59±0.42 <u>ab</u>	11.14±0.10 <u>b</u>	- 0.62±1.14 <u>a</u>	0.71±0.10 <u>ab</u>	5.51 ±0.33 <u>ab</u>
ISSM-N4	304 <u>+22.3a</u>	0.74±0.40 <u>a</u>	12.34±0.16 <u>a</u>	0.55±1.68 <u>a</u>	1.02±0.11 <u>a</u>	6.19±0.63 <u>a</u>
Average 20	11–2013 ^a					
NN ^b	13 <u>5</u> ±19.6d	0.11±0.05c	5.83±0.04f	- 2.08±1.89a	0.48±0.07d	1.75±0.04d
FP ^b	215 <u>±19.9</u> c	0.29±0.13bc	8.59±0.25e	0.22±3.96a	1.45±0.24b	5.67±0.16b
ISSM-N1 ^b	191 <u>+</u> 19. <u>2</u> c	0.18±0.06c	9.45±0.18d	0.42±2.77a	0.84±0.08c	5.04±0.08c
ISSM-N2	192 <u>+</u> 11.6c	0.27±0.12bc	9.98±0.25c	0.15±0.58a	1.08±0.12c	5.76±0.22ab
ISSM-N3	40 <u>2</u> =23.8b	0.50±0.16ab	10.95±0.13b	0.63±3.51a	1.43±0.05b	5.40±0.16bc
ISSM-N4	46 <u>7</u> ±39.2a	0.68±0.15a	12.11±0.28a	2.12±2.57a	1.85±0.16a	6.14±0.35a

^aMean ± 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.

^bSee Table 1 for treatment codes.

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<u>2011–2014 cycle.</u>

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

Crop season	Source	<u>df</u>	<u>CH</u> ₄	<u>N₂O</u>	<u>Yield</u>
_			<u>(kg C ha⁻¹)</u>	<u>(kg N ha⁻¹)</u>	<u>(t ha⁻¹)</u>
<u>Rice</u>	Between subjects				
	<u>P</u>	<u>5</u>	35.3***	3.71***	123***
	Within subjects				
	<u>Y</u>	<u>2</u>	20.7***	0.88**	1.15**
	<u>P×Y</u>	<u>10</u>	6.73***	<u>0.15</u>	<u>0.37</u>
Wheat	Between subjects				
	<u>P</u>	<u>5</u>	<u>0.26</u>	14.8***	76.3***
	Within subjects				
	<u>Y</u>	<u>2</u>	<u>0.55*</u>	15.1***	<u>0.08</u>
	<u>P×Y</u>	<u>10</u>	<u>0.83</u>	4.39***	<u>0.05</u>
Rice-Wheat	Between subjects				
	<u>P</u>	<u>5</u>	<u>37.2***</u>	24.2***	153***
	Within subjects				
	<u>Y</u>	<u>2</u>	20.5***	5.83***	0.70*
	<u>P×Y</u>	10	6.50***	<u>1.11</u>	0.17

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Two-way ANOVA for the effects of cultivation patterns (P) and cropping year (Y) on CH_4 and N_2O emissions, and rice and wheat grain yields for the three annual rice-wheat rotations of 2011–2014. _ Crop season

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.

Agricultural management practices for chemical inputs and farm operations and contributions to carbon dioxide equivalents (kg CO_2 eq. ha⁻¹yr⁻¹) 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	Treatment				Chemical inputs (kg ha ⁻¹) ^a				Farm operations (kg ha ⁻¹) ^c							
Ν	P K	K	K Si	Zn	Herbicide	Insecticide	Fungicide	Irrigation (cm) ^b		Tillage and	Crop	Farm	Crop	Farm machinery		
									2011	2012	2013	raking	planting	manure	harvest	production
NN ^d	0	180	300	0	0	2	18	4	75	80	80	37	1	0	11	135
FP	480	180	300	0	0	2	20	4.4	75	80	80	37	1	0	11	147
ISSM-N1	360	180	300	0	0	2	20	4.4	50	65	55	37	1	0	11	139
ISSM-N2	432	180	300	0	0	2	20	4.4	50	65	55	37	1	0	11	177
ISSM-N3	480	216	360	225	15	2	27	6	50	65	55	37	1	2250	11	177
ISSM-N4	600	252	450	225	15	2	41	9	50	65	55	37	1	2250	11	275
	_				Che	mical inputs (Ei)		_			Farm	operations	(Eo)		
NN	0	132	165	0	0	2	338	53	1419	1514	1514	127	12	0	37	36
FP	2288	132	165	0	0	2	375	59	1419	1514	1514	127	12	0	37	39
ISSM-N1	1716	132	165	0	0	2	375	59	946	1230	1041	127	12	0	37	37
ISSM-N2	2059	132	165	0	0	2	375	59	946	1230	1041	127	12	0	37	47
ISSM-N3	2288	158	198	58	6	2	506	79	946	1230	1041	127	12	62	37	47
ISSM-N4	2860	185	248	58	6	2	768	129	946	1230	1041	127	12	62	37	73

^aThe carbon emission coefficients were 1.3,0.2,0.15, 0.3, 5.1 and 3.9 C cost (kg C eq. kg⁻¹ 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⁻¹ active ingredient) per applied Si and Zn fertilizer, respectively.

^bThe carbon emission coefficient for irrigation was 5.16 C cost (kg C eq. cm⁻¹ ha⁻¹) as referred to in Lal (2004).

^cThe carbon emission coefficients were 0.94, 3.2, 0.0075, 0.94 and 0.0725 C cost (kg C eq. kg^{-1} active ingredient) for tillage and raking, crop planting, per farm manure application, harvesting, spraying and threshing, respectively, as referred to in Lal (2004).

^dSee Table 1 for treatment codes.

reatment	CH ₄	N ₂ O	Ei	Eo	SOCSR	<u>GWP</u> ^a	Grain yield	GHGI ^b
			- kg CO2	eq. $ha^{-1}yr^{-1}$			- t ha ⁻¹ yr ⁻¹	kg CO ₂ eq. t^{-1} grain
NN ^d	4418±628d ^c	276±29d	690	1694	- 792±327c	7871±646d	7.58±0.04d	1038±85b
FP	7181±766c	816±55b	3021	1697	1170±396ab	11545±505c	14.26±0.36c	810±23c
SSM-N1	6381±633c	479±62c	2449	1285	491 <u>+</u> 435b	10104±930c	14.50±0.14c	697±63d
SSM-N2	6421±379c	633±97c	2792	1295	709±193ab	10433±516c	15.74±0.44b	664 <u>+</u> 49d
SSM-N3	13418±744b	906±87b	32 <u>95</u>	1357	1383±503a	175 <u>93</u> ±688b	16.36±0.18b	107 <u>5</u> ±33ab
SSM-N4	15630±1246a	1188±65a	4 <u>256</u>	1383	1545±348a	20 <u>911</u> ±1289a	18.26±0.46a	114 <u>5</u> ±84a
WP (kg CO	$D_2 \text{ eq. ha}^{-1} \text{yr}^{-1}) =$	$25 \times CH_4 + 2$	$298 \times N_2O +$	- Ei +Eo — <u>4</u> 4	<u>I∕12 ×</u> SOCSR, E	Ei (agrochemical in	puts), Eo (farm op	erations), SOCSR (SOC sequestrations)

^cDifferent lower case letters within the same column for each item indicate significant differences at *P*<0.05 based on Tukey's multiple range tests.

^dSee Table 1 for treatment codes.

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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_4) 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 (N2O) 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, 1 Daily mean air temperature and precipitation during the rice-wheat rotation in

2011-2014 in Changshu, China.

已下移 [1]: Fig 1 Daily mean air temperature and precipitation during the rice-wheat rotation in 2011–2014 in Changshu, China.

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