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- 1 MULTIPLE TRADEOFFS BETWEEN MAXIMIZING YIELD AND
- 2 MINIMIZING GREENHOUSE GAS PRODUCTION IN CHINESE RICE
- 3 **CROPLANDS**

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Running title:

27 Tradeoffs between yield and greenhouse gas production

ABSTRACT

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nutrients

30 Globally, paddy fields are a major anthropogenic source of greenhouse gas (GHG) emissions from agriculture. There is, however, limited understanding of relationships 31 32 between GHG production with fertilizer management, rice varieties, and soil variables. This information is crucial for minimizing the climatic impacts of rice agriculture. Here, 33 we examined the relationships between soil GHG production and management practices 34 throughout China. The current doses of N-fertilizer (73-272 kg ha⁻¹) were negatively 35 correlated with rice yield and with CO₂ or CH₄ production, and positively correlated with 36 N₂O production, thus suggesting N-overfertilization. Impacts on soil traits such as 37 38 decreasing pH or the availabilities of other nutrients could be underlying these relationships. Rice yield was highest and GHG production was lowest at sites using 39 40 intermediate levels of P- and K-fertilization. CO₂ and CH₄ production and emissions were 41 positively related with soil water content. The yield was higher and N₂O productions were lower at the sites with japonica rice. Our results strongly suggest that current high doses 42 43 of N-fertilizers could be reduced to thus avoid the negative effects of excessive N input on GHG production without any immediate risk of rice production loss. Current 44 intermediate doses of P- and K-fertilization should be adopted across China to further 45 46 improve rice production without the risk of GHG emissions. The use of different rice varieties and strategies of water management should be re-examined in relation to crop 47 production and GHG mitigation. 48 KEYWORDS: paddy field, greenhouse gases, yields, nitrogen, phosphorus, soil 49

1. INTRODUCTION

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52 Rice currently feeds more than 50% of the global population (Haque, Kim, Ali & Kim, 2015), but production will need to increase by 40% by the end of 2030 worldwide to meet 53 54 the demand for food from the growing population (FAO, 2009). Sustaining soil fertility and increasing rice yields are therefore of utmost importance. An increased nutrient 55 supply can stimulate the growth and grain yield of rice plants (Ali, Oh & Kim, 2008; 56 57 Wang et al., 2014) but can also influence the potential of paddy fields to produce and emit 58 greenhouse gases (GHG). Globally, paddy fields are a major anthropogenic source of greenhouse gas (GHG) emissions from agriculture (Tan, 2011). Paddy fields are very 59 important sources of GHG, especially methane (CH₄) and nitrous oxide (N₂O) (Myhre et 60 al., 2013), so minimizing the release of these very potent GHG could contribute to 61 62 mitigating their adverse impacts on climate change (Li et al., 2006). Chinese GHG emissions from agricultural systems account for ~ 40% of Chinese GHG emissions, hence 63 requiring detailed investigations. Furthermore, since sixty percent of the Chinese 64 65 population depends on rice-based food, so protecting China's rice production for food 66 security is important (Zhu, 2006). There is, however, limited understanding of relationships between GHG production 67 68 with fertilizer management, rice varieties, and soil variables. This information is crucial for minimizing the climatic impacts of rice agriculture, especially for the agricultural 69 70 sustainable development in China. Improving the status of soil nutrients in paddy fields 71 for improved rice yield while decreasing GHG emissions, or at least not increasing it, is a challenging option. However, intense fertilization can also induce rises in GHG 72 emissions in paddy soils (Fan et al., 2016), and great soil nutrient concentration also can 73 74 be related to GHG emissions in paddy soils (Wang et al., 2017a). Rice crops in China are frequently overfertilized (Cheng &Li, 2007) so a general analysis of the relationships of 75

fertilization and soil traits with GHG emissions and crop yield is necessary to detect the level of over-fertilization, GHG emissions and yield, and moreover, to improve the future management of the sustainable development of rice agriculture. Here, we examined the relationships between soil GHG production and management practices throughout China.

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Most current studies associating GHG emissions from paddy fields with soil properties have been conducted at a single location by applying different treatments that modify soil properties (Wassmann et al., 1998; Wang et al., 2014). Several strategies for managing rice crops, such as from fertilizer and herbicide application to straw or water management, are aiming to increase rice production. However, such management strategies may also increase or decrease CO₂, CH₄ and/or N₂O emissions (Li et al., 2005, 2013; Liu et al., 2016; Jiang, Chen, Sun, Song & Huang, 2015; Launio, Asis, Manalili & Javier, 2016; Trinh et al., 2017; Wang et al., 2016c; Zhang et al., 2016a,b; Jiang, Chen, Sun, Song & Huang, 2015; Launio, Asis, Manalili & Javier, 2016; Trinh et al., 2017; Wang et al., 2016c; Zhang et al., 2016a,b). Many of these studies have reported links between GHG emissions and various soil traits, such as pH (Wang et al., 2017a), redox potential, (Fan et al., 2016; Wang et al., 2017b), salinity (Olsson et al., 2015), sulfate concentration (Dong et al., 2011; Theint, Susuki, Ono & Bellingrath-Kimura, 2014; Wang et al., 2017b), N content (Zhu, Zhang & Cai, 2011; Zheng, Zhang & Cai, 2013; Zhao et al., 2015; Wang et al., 2017c) and soil P concentration (Adhya, Pattnaik, Satpathy, Kumaraswamy & Sethunathan, 1998; Zheng, Zhang & Cai, 2013; Sheng et al., 2016). However, only few studies have tested the differences in GHG productions and emissions at sites with different soil traits and management strategies, including fertilization. Studying soil conditions on a large scale, including coastal and inland paddy soils at large regional scales, and their relationships with GHG productions and emissions are thus warranted for increasing rice production while controlling GHG emissions.

We hypothesized that different fertilization practices, strategies of rice-crop management, rice varieties, concentrations of soil carbon (C) and other nutrients, salinity or/and pH would explain a large part of the differences in GHG emissions and yield among sites, especially, the fertilization amount increment will increase the GHG emissions but not the yield. Our results will provide information for improving strategies and managing soil conditions toward more favorable traits to avoid a possible increase in GHG production without yield loss. We pursued this objective by determining (1) the relationships among fertilization dose, soil GHG production and rice yield in China, (2) the relationships among GHG production, rice variety and environmental traits, such as region (sites), location (coastal vs. inland) and cropping system (single or double) on these relationships.

2. MATERIALS AND METHODS

113 2.1 Study area

This study was conducted throughout China (Figure 1). China has 2.45×10^7 ha of cultivated rice, and 90% of the paddies are in the subtropics. China has a large area of paddy rice, diverse soil types and different tillage and fertilizer management practices, all of which may affect the content and distribution of nutrients and GHG emissions. We collected the soils in the whole China rice cultivation areas choosing sites with contrasting fertilization management. The studied ranges of fertilization were 73 -272 kg ha⁻¹ for N fertilizer, 48-150 kg ha⁻¹ for P₂O₅, and 45-270 kg ha⁻¹ for K₂O. The main characteristics of the different studied sites are showed in Table S1. To analyze the role of sea proximity we separated the studied sites between inland and coastal rice crops depending on the distance to sea line; when less than 20 km was considered coastal paddy field and more than 20 km was considered inland paddy field. To analyze the role of fertilization intensity we classified the distinct sites as low, intermediate and high intensity fertilized. In the

case of N-fertilization: Low <100 kg N ha⁻¹ y⁻¹, intermediate between 100-150 kg N ha⁻¹ y⁻¹ and high intensity >150 kg N ha⁻¹ y⁻¹. In the case of P-fertilization: Low<60 kg P₂O₅

 $128~ha^{\text{-}1}~y^{\text{-}1}$, intermediate between 60-75 kg $P_2O_5~ha^{\text{-}1}~y^{\text{-}1}$ and high intensity>75 kg $P_2O_5~ha^{\text{-}1}$

y⁻¹. In the case of K-fertilization: Low <60 kg K₂O ha⁻¹ y⁻¹, intermediate between 60-70

130 kg K_2O ha⁻¹ y⁻¹ and high intensity >70 kg K_2O ha⁻¹ y⁻¹.

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2.2 Collection and measurement of soil samples

Soil samples were collected from 34 randomly selected paddy fields and five replicated plots throughout China using a small core sampler (length and diameter of 0.5 and 0.1 m, respectively) from the plowed (0-15 cm) soil layer in October to November 2015 (Figure 1). A total of 170 samples (34 sites \times 1 soil layer \times 5 replicates) were thus collected. We attributed China into six regions for this study: Northeast China, North China, East China, Center and South China, Southwest China and Northwest China (Sun, Huang, Zhang & Yu, 2010). Moreover, the sampling site characteristics and paddy field management were also investigated. The collected soil samples were also used for all analyses. The samples were air-dried, and roots and visible plant debris were removed. Total C and N contents were measured using a Vario EL III Elemental Analyzer (Elementar Scientific Instruments, Hanau, Germany, Wang et al., 2015a; Wang et al., 2016a,b). Labile organiccarbon (LOC) content was determined by digestion with 333 mM KMnO₄ (Wang, Lai, Wang, Oan & Zheng, 2015b), while NH₄⁺ and NO₃⁻ were determined by extracting the soils with 2 M KCl (Lu, 1999). Available N content was the sum of NH₄⁺ and NO₃⁻. The total P content was determined, first by perchloric-acid digestion and then using a sequence flow analyzer (San++, SKALAR Corporation production, Breda, The Netherlands). To determine soil P availability, we used the Mehlich extraction method and then using a sequence flow analyzer (San++, SKALAR Corporation production,

Breda, The Netherlands).

The soil CO₂, CH₄ and N₂O productions were determined using anaerobic incubation consistent with soils under water saturation (Wang et al., 2015a). Thirty grams of fresh soil of the five core samples for each site were placed in 120-mL incubation bottles, and two volumes of distilled water were added. The bottles were purged with N₂ for 2 min to replace the O₂ and were then sealed with a rubber stopper and incubated at 25 °C for 3 d. Five milliliters of gas were extracted from the headspaces each day, about 24 h interval in four times during incubation: 0, 24, 48 and 72 hours during incubation experiments. This method has been successfully used in several previous studies (Wassmann et al., 1998; Wang et al., 2017b). We also use the method, of Xu et al., 2016 to calculate annual gas emissions from some days (in our case 3 days) of sampling (Xu et al., 2016).

The soil CO₂ and CH₄ concentrations in the samples of headspace air were determined by gas chromatography (Shimadzu GC-2010, Kyoto, Japan). The soil N₂O concentrations in these samples were determined by gas chromatography (Shimadzu GC-2014, Kyoto, Japan) using a stainless-steel Porapak Q column (2 m length, 4 mm OD, 80/100 mesh). A methane conversion furnace, flame ionization detector (FID) and electron capture detector (ECD) were used for determining the CO₂, CH₄ and N₂O concentrations, respectively. The operating temperatures of the column, injector and detector were adjusted to 45, 100 and 280 °C, respectively, for determining the CO₂ concentrations; to 70, 200 and 200 °C, respectively, for determining the CH₄ concentrations; and to 70, 200 and 320 °C, respectively, for determining the N₂O concentrations. Helium (99.999% purity) was used as a carrier gas (30 mL min⁻¹), and a make-up gas (95% argon and 5% CH₄) was used for the ECD. The gas chromatograph was calibrated before and after each set of measurements using 503, 1030 and 2980 μL

 $CO_2 L^{-1}$ in He; 1.01, 7.99 and 50.5 μ L $CH_4 L^{-1}$ in He and 0.2, 0.6 and 1.0 μ L $N_2O L^{-1}$ in

177 He (CRM/RM information center of China) as primary standards (Wang et al., 2015c, d).

We used linear equations for calculating CO₂, CH₄ and N₂O productions.

Other soil variables were also analyzed. Bulk density was measured using three 15 × 3 cm cores (Wang et al., 2016b), and was estimated by core mass dry weight divided by core volume, and represent the averaged bulk density of 0-15 cm. Soil water content was measured by the drying method (Lu, 1999). pH was measured with a PHS-3C pH meter (Orion Scientific Instruments, Minnesota, USA) and salinity was measured using a 2265FS EC Meter (Spectrum Technologies Inc., Paxinos, USA; Wang *et al.*, 2016b). Soil particle size (percent clay, silt and sand contents) was measured by a Mastersizer 2000 laser particle-size analyzer (Malvern Scientific Instruments, Malvern, UK; Wang *et al.*, 2016b).

2.3 Determination of soil C and nutrient contents

191 The total C, N and P contents, and labile organic carbon (LOC), available N, available P,

NH₄⁺-N and NO₃⁻-N contents in the 0-15 cm soil profile were estimated by following the

approach of Mishra, Ussiri & Lal (2010):

$$C_{\rm S} = \sum C_{\rm m} \times \rho_{\rm b} \times D$$

where C_S is the total C, N or P content or LOC, available N, available P, NH₄⁺-N or NO₃⁻

-N content (kg m⁻²), C_m is the total C, N or P content or LOC, available N, available P,

 NH_4^+ -N or NO_3^- -N content (g kg⁻¹), ρ_b is the bulk density (kg m⁻³), D is the thickness of

each soil layer (0.15 m).

The total C, N and P contents, and LOC, available N, available P, NH₄⁺-N and NO₃⁻

-N contents were calculated for each region by multiplying area content and areas of each paddy distribution. The contents for each measured variables across China were calculated as the sum for all regions.

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- 2.4 Determination of soil CO₂, CH₄ and N₂O productions
- We used linear regressions for calculating soil CO₂, CH₄ and N₂O productions
- 207 (Wassmann *et al.*, 1998):

$$P = \frac{dc}{dt} \cdot \frac{V_H}{W_S} \cdot \frac{MW}{MV} \cdot \frac{T_{st}}{T_{st} + T}$$

- where P is the rate of CO₂, CH₄ or N₂O production ($\mu g^{-1} g^{-1} d^{-1}$), dc/dt is the recorded
- change in the mixing ratio of CO₂, CH₄ and N₂O in the headspace over time (mmol mol
- 211 1 d⁻¹), V_H is the volume of the headspace (L), W_S is the dry weight of the soil (g), MW is
- the molecular weight of CO_2 , CH_4 or $N_2O(g)$, MV is the molecular volume (L), T is the
- temperature (K) and T_{st} is the standard temperature (K).
- The potential productions of CO₂, CH₄ and N₂O, calculated as the average
- 215 productions per unit area per year [productions rate × soil depth × bulk density × days of
- 216 the year (365 d in 2015)]. The productions of CO₂, CH₄ and N₂O were calculated for each
- 217 region as average production per unit area per year × area of the paddy fields in the
- 218 production region. The GHG productions for all of China was calculated as the sum of
- 219 productions form all paddy regions.

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- 221 2.5 Statistical analyses
- One-way ANOVAs with N-, P- and K-fertilization doses categorical (low, intermediate
- and high intensity), regions, inland-coastal environments and cropping systems as
- independent variables and gas production variables, and yield as a dependent continuous

variable were conducted with Bonferroni post hoc tests. The data were checked for normality and homogeneity of variance, and if necessary, were log-transformed. We used the Benjamini–Hochberg procedure to control the rate of false discovery (Benjamini and Hochberg 1995) to analyze the relationships between gas emissions and all the studied environmental soil and climate variables. The different individual analyses were listed in rank order according with their ascending P-value. Thereafter, for each single analyses each P-value was divided by the total number of test and thereafter multiplied by the false discovery rate (habitually 0.25) then the values below 0.05 were considered as significant. We also used Tukey's method (Tukey, 1977) to detect and remove outliers.

We also performed multivariate statistical analyses. We used principal component analyses (PCAs) to determine the overall differences of the soil variables and CO₂, CH₄ and N₂O productions rates, and annual accumulated emissions among fertilizer doses. We used all variables given the scarce multicollinearity existing among variables (see Table S1), with no $R^2>0.6$ between any pairwise variables. We have also estimated VIF for each independent fixed variables in the mixed models. We conducted one-way ANOVAs with Bonferroni post hoc tests of the scores of the first PC axis to determine differences among the treatments. We then used general discriminant analyses (GDAs) to determine the overall differences of soil traits; CO₂, CH₄ and N₂O productions rates and annual accumulated emissions and yield among fertilizer doses, and productivities among sites with different yields. Discriminant analyses consist of a supervised statistical algorithm that derives an optimal separation between groups established a priori by maximizing between-group variance while minimizing within-group variance (Raamsdonk et al., 2001). GDA is thus an appropriate tool for identifying the variables most responsible for differences among groups. The GDAs and PCAs were performed using Statistica 8.0 (StatSoft, Inc., Tulsa, USA). Before conducting these multivariate analyses, we selected

the sampling adequacy of individuals and the set of variables by the Barlett's test of spherity (<0.05) and the Kaiser-Meyer-Olkin measure (>0.50). We removed the variables with communality values < 0.5 and perform the PCA and DGA analyses with the variables with communality > 0.5. To perform these sampling adequacy analyses we used the package psych (Revelle, 2010).

Significant differences in CO₂, CH₄ and N₂O productions among does of fertilizers, number of crops, crop location (inland vs. coastal) and soil traits were tested by general mixed models using location with topography, site and plot as random nested factors. We used the "Ime" function of the "nlme" R package (Pinheiro, Bates, DebRoy, Sakar & Core, 2016). Non-normally distributed variables were log-transformed. We chose the best model for each dependent variable using the Akaike information criterion (AIC). We used the MuMIn (Barton, 2012) R package in the mixed models to estimate the percentage of the variance explained by the model. We conducted Tukey's post hoc tests to detect significant differences in the analyses for more than two communities using the "multcomp" (Hothorn, Pretz & Wersfall, 2013) R package with the "glht" function. The relationships of each soil variable with CO₂, CH₄ and N₂O productions were determined by simple regressions using Statistica 6.0 (StatSoft, Inc. Tulsa, USA). We used the Benjamini–Hochberg procedure to control for rates of false discovery (Benjamini & Hochberg, 1995).

We used structural equation modeling (SEM) to study the total effects of fertilizer doses on accumulated gas emissions by both direct and indirect effects of soil traits. We fit the different models using the SEM R package (Fox, Nie & Byrnes, 2013) and determined the minimum adequate model using AIC. Standard errors and significance levels (P values) of the total, direct and indirect effects were calculated using bootstrapping (1200 repetitions) (Davison, Hinkley & Schechtman, 1986; Mitchell-Olds,

275 1986).

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3. RESULTS

3.1 Effects of fertilization doses, regions, rice variety, inland-coastal environments and 278 cropping systems on soil GHG productions and rice yield 279 One-way ANOVAs with N-, P- and K-fertilization doses as independent categorical (low, 280 281 intermediate and high intensity) variables and gas production variables, and yield as a dependent continuous variable, indicated that the lowest levels of N fertilization (<100 282 kg N ha⁻¹ y⁻¹) were associated with the highest annual yields. However, the highest levels 283 of N fertilization (>150 kg N ha⁻¹ y⁻¹) were associated with the lowest annual yields 284 (Figures 2d and S1). The lowest doses of N fertilizer were associated with the highest soil 285 286 CO₂ and CH₄ productions, and the lowest N₂O productions, with the opposite patterns at the highest doses of N fertilizer. The intermediate doses of P (60-75 kg P₂O₅ ha⁻¹ y⁻¹) and 287 K (60-70 kg K₂O ha⁻¹ y⁻¹) fertilizers were the best, because they were associated with the 288 289 highest yields and the lowest CO₂ and CH₄ productions (Figures 3a,b, S2, 4a,b and S3). 290 The correlations among all the studied variables were shown in Table S2.

The balance between yield and GHG production was worst in the East China rice crops, with the highest productions of CO₂, CH₄ and N₂O and the lowest annual yield (Figures 5, and S4). Coastal rice crops have higher CO₂ and CH₄ production (Figure 6a,b, S5a,b) and lower yield than inland rice crops (Figure 6d). Total annual yield was notably similar at the sites with one and two annual rice crops (Figure 7d). The CO₂ production rates were higher at sites with one annual crop than at sites with two annual crops (Figure S6a,c).

We excluded hybrid and glutinous rice varieties from the analysis of the effects of rice variety, because both were only at one site each. We focused on japonica and Hsien

rice varieties, which were the main varieties planted at the sites. On average, the yield was higher and N₂O productions were lower at the sites with japonica rice (Figure S7).

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3.2 Gas productions and environmental traits

We observed scarce relationships with studied gas emissions and production with the studied soil variables, the most strong was the positive relationships between total soil N concentrations and CO₂ emission and production and between soil water content and methane emission and production (Table S2). The variables that mainly loaded on PC1 (correlation >0.4) were Soil N concentration, LOC soil extractable NH4+ and NO3concentrations, soil pH, soil salinity, soil bulk density, sand %, MAP, soil NO₃ content, CO₂ emission rates and yield. The variables that mainly loaded PC1 were soil total C, N and P concentrations, soil P availability, soil NO₃⁻ concentrations and contents, and soil salinity. A PCA of all available data also indicated that the rice crops under the low to intermediate doses of N fertilizer, and intermediate doses of P and K fertilizers had the highest yields and lower CO₂ and CH₄ productions (Figure 8a). Whereas, crops receiving high doses of N and K fertilizers had the lowest annual yields and the highest CO₂ and CH₄ productions, as shown along the PC1 axis (Figure 8b). The PCAs indicated that the North China and Northwest China rice crops were plotted by the two first PC axes toward higher yield and soil P availability, total P content, C:N ratio and sand content, and toward lower soil N:P ratio, labile C:N ratio and labile C:P ratio and lower CO2 and CH4 productions (Figure 8b). Fields in the East China, Center and South China, Southwest China regions had opposite patterns.

Coastal rice crops have higher CO₂ and CH₄ emissions and lower yield than inland rice crops. This was associated with higher soil N, water and clay contents in coastal rice crops (Figure 8c). Figure 8d shows the overall differences in the environmental variables

between the sites with one and two annual crops. These two groups of sites were clearly separated along the PC1 axis, with single-crop sites plotted toward higher annual yields and soil P contents, salinities, C:N ratios and pHs, whereas the double-crop sites were plotted toward higher soil N, water and LOC contents and N:P ratios, and higher CO₂ productions.

The best mixed models (based on a low AIC and the highest R² and parsimoniousity), with rice variety and environmental traits as fixed independent variables; location, topography, site and plot as random factors; and GHG productions as dependent variables, indicated that 26% of the total variance of annual accumulated CO₂ productions was explained by the length of the growth period, soil water content and bulk density (Table S3). Thirty-nine percent of the total variance of CO₂ productions rates was explained by the growth-period length, soil water content and P-fertilizer dose. Forty-seven and 50% of the total variance of annual accumulated CH₄ productions and CH₄ productions rate, respectively, were explained by the growth-period length, soil water content, water source (river or groundwater), rice variety, bulk density, total soil N content and P-fertilizer dose. Nineteen and 21% of the total variance of annual accumulated N₂O productions and N₂O productions rate, respectively, were explained by the cropping systems, soil P availability and soil NO₃-N content. All six mixed models (Table S3), corresponding to each of the production variables, explained >99% of the corresponding total variance, while taking into account the random variables.

3.3 Relationships between annual yield and gas productions; fertilization effects

The GDA indicated that the crop sites with high yields generally had the highest GHG productions, whereas crop sites with moderate yields generally had the lowest GHG productions (Figure S8). Lower doses of N fertilizer were surprisingly associated with

higher yields but also with higher CH₄ productions (Figure S9), whereas intermediate doses of N fertilizer were associate with higher N₂O productions, consistent with the above results. Intermediate doses of P fertilizer were associated with the highest annual yields and the lowest CO₂ and CH₄ productions (Figures S10), whereas intermediate doses of K fertilizer were associated with the highest annual yields and the lowest CH₄ productions (Figures S11) also consistent with the results of the one-way ANOVAs.

When the productivities at the various doses of N, P and K fertilizers (yield kg⁻¹ fertilizer) were used as grouping dependent factors in the GDA, the sites with high N productivity were associated with the highest N₂O productions, sites with intermediate N productivity were associated with the highest CH₄ productions, and sites with low N productivity were associated with higher CO₂ productions (Figure S12). The lowest CO₂, CH₄ and N₂O productions were associated with sites with intermediate P fertilization (Figure S13), and sites with intermediate K fertilization were associated with the lowest CO₂ and CH₄ but not N₂O productions (Figure S14).

3.4.SEM models

The SEM provided further evidence of the complex relationships among fertilizer doses, GHG productions and annual yield. The N fertilization had a positive direct effect on GHG productions and a negative effect on annual yield, and P fertilization had the opposite pattern (Figure 9), and K fertilization did not significantly affect GHG productions but generally negatively affected yield (Figure 9c). The N fertilization also had indirect negative effects on annual yield by increasing the amount of labile soil carbon content and by lowering soil pH. We also detected a negative direct and indirect effect, by decreasing soil water content, effect of bulk density on soil CO₂ productions (Figure 9b).

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4. DISCUSSION

We studied potential emissions of carbon dioxide (CO₂), CH₄ and N₂O from paddy fields across China, because they are the most important GHG, contributing about 80% of the current global radiative forcing (Myhre et al., 2013). The statistical analyses of our database of 34 field sites throughout China clearly identified a general trend to over fertilization with N. The current dose of N fertilizer was negatively correlated with rice yield. This correlation was exacerbated by a positive correlation between N-fertilization and N₂O productions and negative correlations between N-fertilization and CO₂ and CH₄ productions. These results are generally consistent with previous findings, that have also observed that higher N application doses had detrimential effects on yield and/or increased gas emissions. Kim et al. (2016a) reported that rice yield increased with Nfertilizer dose to levels of 110-130 kg N ha⁻¹ y⁻¹ and thereafter decreased at higher levels. A meta-analysis of 24 field studies of rice crops in China demonstrated that N fertilization increased CH₄ and N₂O emissions and decreased the ratio of yield-to-GHG emission, which was highest for 150-200 kg N ha⁻¹ y⁻¹ (Feng et al., 2013) very similar results than the observed in our study. Zhong, Wang, Yan & Zhao (2017) observed that the percentage of N lost from leaching, ammonia volatilization and denitrification increased with the Nfertilizer dose in a field experiment in China. Our results also provide evidence that both increased soil N and P availability increased rice yield (Figure S4), even though the Nfertilizer dose was negatively correlated with yield. These results supported the hypothesis that increasing the doses of N fertilizer can have more detrimental than positive effects on yield and GHG productions, even though the availability of N to plants is important and positive for rice production. The results

thus strongly suggest a possible N saturation and/or ineffective N fertilization beyond a certain level, with no extra positive effects but mostly negative effects of the higher N input on rice production. Current reports, however, demonstrate an interest to optimize N-fertilization doses in rice crops. Zhu, Zhang, Zhang, Deng & Zhang (2016) reported a decrease in GHG emissions and an increase in rice yield as N-fertilizer doses decreased and plant density increased (~50% increase) in northeastern China. Zhang *et al.* (2016b) similarly demonstrated that the doses of N fertilizer typical of Nanjing paddy fields (China) could be decreased without decreasing rice yield but could decrease GHG emission.

Different studies have reported contradictory effects of N fertilization on GHG emissions from rice crops (see the review by Cai, Shan & Xu, 2007). Several studies, however, have observed an increase in N₂O emissions with increases in N-fertilizer dose in rice croplands (Zhang *et al.*, 2014; Kim, Jeong, Kim, Kim & Kim, 2016b).

Our data analyses indicated that P- and K-fertilizer doses generally had positive effects on rice yield and decreased GHG productions. The relationships among P- and mainly K-fertilization, yield and GHG productions in rice croplands have not been studied much, as compared to the corresponding relationships with N-fertilization. Our results are nonetheless consistent with other experimental studies (Li et al., 2013; Datta, Santra & Adhya, 2013). Datta, Santra & Adhya (2013) found that N fertilization explained more of the changes in CH₄ emissions than P or K fertilization. In contrast, Li et al. (2013) observed that increasing the dose of P fertilizer decreased CH₄ and N₂O emissions but increased yield.

However, our study has some potential limitations. The incubation period of three days could not be sufficient to capture the overall patterns of GHG production and emission, and thus has limited reflection of the field condition, but it is still very useful

to compare the potential emission among studied sites. In the paddy fields, the first peak of methane emission generally occurs within a month after transplanting, just according with our study. But a second peak would occur at approximately two months and this is mainly governed by the stable low soil redox potential and neutral soil pH, and the increased release of plant-borne carbon sources. (e.g. Ly et al., 2013; Vu et al., 2015). This can be partially corrected by using Xu et al. 2016 method to estimate all year gas emissions. But all in all this can explain why the estimated methane production in this paper is lower than in other reports in Asian countries (Yan et al., 2003; Vo et al., 2018).

4.1. Regions, locations and cropping systems

Our results suggest that rice yield can be increased without increasing or even decreasing GHG productions. For example, our results indicated that yields were higher for the japonica than the Hsien varieties. The N₂O productions were several times higher for the Hsien varieties, leading to a better balance between yield and GHG productions in fields with japonica varieties, even though CH₄ productions were clearly higher for the japonica varieties. Adequate irrigation is fundamental for assuring high rice yield (Sun et al., 2016), but our data also suggest that GHG productions can be reduced by regulating doses of N-, P-, and/or K-fertilization. We have also observed that CH₄ and N₂O productions were much higher at sites irrigated and flooded with river water than at sites irrigated and flooded with groundwater or water from superficial reservoirs, whereas average yield did not differ significantly between these sites. This result is difficult to interpret in the context of this study; further studies should aim to find out the cause of these differences. The balance between yield and GHG production was worst in the East China rice crops, with the highest productions of CO₂, CH₄ and N₂O because in this area the temperature is relatively higher, and there are more active substrates (Wang et al., 2015). The North

China and Northwest China rice crops had the opposite patterns because in this area the lower temperature limits the substrates decomposition, such as soil organic carbon, and then the microbes act to convert plant residues into humus in the soil (Cui et al., 2008). The lower temperature can decrease decomposition and the CO₂ and CH₄ release from soil by mediating the microbe growth (Tang et al., 2017).

Moreover, in these areas paddy soils had relative higher pH and water comes from rivers that can provide more substrate to the paddy and also have longer growth period and more illumination than in other areas of china. Furthermore, the North China and Northwest China rice crops lower greenhouse gases production was related with the higher C:N ratio. The C:N ratio controls the CO₂ and CH₄ release. There is more limited carbon decomposition with relatively higher C:N ratios (Windham, 2001). The soil GHG productions tended to be lower (significantly for CO₂) at inland than coastal sites because he coastal paddy fields had higher carbon and nitrogen concentration, and therefore more substrates for soil GHG productions (Delaune et al., 2018).

Moreover, the CO₂ production rates were higher at sites with one annual crop than at sites with two annual crops because the two annual areas, mostly in the south of China, had soils rich in ferric oxide, which, in turn, favored C fixation in soil, and more stable soil carbon, thus decreasing C release in form of methane (Wang et al., 2014). Furthermore, CH₄ production was higher under single crop. Single crop areas had lower temperature during the whole year, thus lowering decomposition, increasing carbon storage in the soil and providing more substrates for CH₄ production when the temperature increases during rice growth period. However, N₂O is higher under double crops because of more applications of N fertilizer.

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storage in the soil and more substrates is able for CH₄ production when the temperature increase during rice growth period. However, N₂O is higher under double crops, because along the year the N fertilizer applied is in higher amount in the doubles crop areas.

Our multivariate analysis of all data from all sites indicated that the highest average yields were accompanied by the highest GHG productions. Optimizing rice production and GHG production is thus interesting and challenging. Our analyses provide some clues for optimization, suggesting that some advances could be achieved by adjusting N-fertilization to a level that improve rice production but avoid the negative effects of excessive N input. Further, GHG productions could be reduced by maintaining adequate levels of P and K fertilizers, mostly at intermediate doses of the current range of P and K fertilization across China, and by re-examining the use of different rice varieties and strategies of water management.

5. CONCLUSION

Rice production has historically been improved using N fertilizers, but we found that the current paddy field sites in China with relatively low N fertilization had high rice production and low soil CO₂ and CH₄ potential productions, even though rice yield was positively correlated with soil N availability. In contrast to N fertilization, sites using intermediate doses of P and K fertilizers had the highest rice yields and the lowest soil GHG potential productions. The large capacity of soil to accumulate P in non-available forms and the large capacity of K leaching by the high use of water in rice crops together with a less direct role of P and K in CH₄ and N₂O productions could explain these results.

The analysis of all our data strongly suggests that increasing rice yield and minimizing GHG productions can be further optimized in China. The use of different rice varieties and strategies of water management and fertilization with implications for

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Figures

- Figure 1. Locations of the field sites for soil sampling across China.
- Figure 2. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O for the N-
- fertilizer annual doses (low < 96 kg ha⁻¹, intermediate 96-140 kg ha⁻¹, high more than 140
- kg ha⁻¹). Different letters indicate significant statistical differences (P<0.05).
- Figure 3. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O for the P-
- fertilization annual doses (low < 62 kg ha⁻¹, intermediate 72-92 kg ha⁻¹, high more than
- 773 72 kg ha⁻¹). Different letters indicate significant statistical differences (P<0.05).
- Figure 4. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O for the K-
- fertilizer annual doses (low < 70 kg ha⁻¹, intermediate 71-81 kg ha⁻¹, high more than 81
- kg ha⁻¹). Different letters indicate significant statistical differences (P<0.05).
- Figure 5. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O in the
- various regions. Different letters indicate significant statistical differences (P<0.05).
- Figure 6. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O at inland
- vs coastal sites. Different letters indicate significant statistical differences (P<0.05).
- Figure 7. Rice yield and total accumulated productions of CO₂, CH₄ and N₂O at sites
- 782 with one vs two rice crops. Different letters indicate significant statistical differences
- 783 (P<0.05). Different letters between brackets indicate marginal significant statistical
- 784 differences (P<0.1).
- Figure 8. First two PC axes of the PCA of all soil variables, GHG productions and yield
- showing the areas (95% confidence intervals) occupied by sites with the N-, P- and K-
- fertilizer doses (A), the areas (95% confidence intervals) occupied by sites in the various
- regions (B), showing the areas (95% confidence intervals) occupied by sites for the inland

vs coastal locations (C) and showing the areas (95% confidence intervals) occupied by sites with one vs two rice crops (D). The Acronyms mean: CO₂ rate= CO₂ productions rate, CH₄ rate= CH₄ productions rate, N₂O rate= N₂O productions rate, [C]=Soil C concentration, [N] = Soil N concentration, [P] = Soil P concentration, NH₄= Soil NH₄⁺ concentration, NO₃= Soil NO₃⁻ concentration, NO₃cont= Soil NO₃⁻ content, LOC= Soil labile organic carbon concentration, P avai= Total soil available P concentration, Sand=Percentage in weight of sand in dry soil, Clay= Percentage in weight of clay in dry soil, pH=Soil pH, Water=Soil water content, salinity=Soil salinity, Bulkd=Soil bulk density, Yield=Rice crop yield, MAP=Mean Annual Precipitation, MAT=Mean Annual Temperature.

Figure 9. SEM models with soil CO₂ production rate (A), soil accumulate CO₂ production (B) and rice crop yield (C) as end endogen variables. Numbers on the arrows are the estimates of the effects of one variable (near the beginning of the arrow) over another (near the end of the arrow) and the corresponding *P* values (in parentheses). Red arrows indicate negative relationships, and black arrows indicate positive relationships.