

1 **Rice straw incorporation affects global warming potential differently**  
2 **in early vs late cropping seasons in southeastern China**

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20 **Abstract**

21 Paddy fields are a major global anthropogenic source of methane (CH<sub>4</sub>) and nitrous  
22 oxide (N<sub>2</sub>O), which are very potent greenhouse gases. China has the second largest  
23 area under rice cultivation, so developing valid and reliable methods for reducing  
24 emissions of greenhouse gases while sustaining crop productivity in paddy fields is of  
25 paramount importance. We examined the effects of applying straw, a residual product  
26 of rice cultivation containing high amounts of carbon and nutrients, to rice crops  
27 during both an early crop season (5 April - 25 July 2012) and a late crop season (1  
28 August - 6 November 2012) on CH<sub>4</sub> and N<sub>2</sub>O emissions in a subtropical paddy field  
29 in southeastern China. CH<sub>4</sub> fluxes had two seasonal peaks, on 5 May and 28 June, in  
30 the early crop but only one peak, on 13 August, in the late crop, which could be  
31 attributed to the lower temperatures after the final tillering stage in the late crop.  
32 Straw application significantly increased mean CH<sub>4</sub> cumulative production (g m<sup>-2</sup>)  
33 relative to the control in the late crop (37.3 vs. 8.34 mg m<sup>-2</sup>, *P*<0.05) but not in the  
34 early crop (0.83 vs. 01.13 mg m<sup>-2</sup>, *P*>0.05). The application of straw significantly  
35 increased N<sub>2</sub>O cumulative production relative to the control in the late crop (75.9 vs.  
36 43.4 μg m<sup>-2</sup> h<sup>-1</sup>) but decreased N<sub>2</sub>O cumulative production by over 43% in the early  
37 crop (15.60 vs. 27.27 μg m<sup>-2</sup> h<sup>-1</sup>) (*P*<0.05). Straw application increased rice yield by  
38 9.63% and 12.58% in early and late crop respectively. Straw incorporation decreased  
39 global warming potential in the early season, but increased it in the late season. Thus,  
40 despite straw application enhances emissions of greenhouse gases in some situations,  
41 its application in the adequate season (here early crop) may be an effective soil

42 amendment that can increase soil fertility without enhancing or even mitigating  
43 emissions of greenhouse gases and thus climate change.

44 **Keywords:** Rice paddy; CH<sub>4</sub> flux; N<sub>2</sub>O flux; Straw application; Seasonal variation

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46 **Highlights**

- 47 • Straw application had no significant effect on CH<sub>4</sub> flux in the early crop
- 48 • Straw application reduced N<sub>2</sub>O flux by over 43% in the early crop
- 49 • Straw application significantly increased CH<sub>4</sub> and N<sub>2</sub>O fluxes in the late crop
- 50 • The lower temperatures during late crop were related with the low CH<sub>4</sub>
- 51 emissions.

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53 **1. Introduction**

54 Anthropogenic emissions of greenhouse gases (GHGs) and the associated climate  
55 change are major global environmental problems in the 21<sup>st</sup> century. Agricultural  
56 activities are responsible for ca. 20% of the current concentrations of GHGs in the  
57 atmosphere (Hütsch, 2001). Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) are two of the  
58 most important GHGs, with global warming potentials (GWPs) of 28 and 265,  
59 respectively, relative to carbon dioxide over a 100-year period (Myhre et al., 2013).  
60 The rapid increases in the atmospheric mixing ratios of CH<sub>4</sub> and N<sub>2</sub>O from  
61 preindustrial levels of 722 and 270 ppb to the current levels of 1830 and 324 ppb,  
62 respectively (Myhre et al., 2013), have created an urgent need to reduce GHG  
63 emissions to the atmosphere to mitigate the adverse impacts of climate change.

64 Over 70% of the global CH<sub>4</sub> emissions originate from biogenic sources (Denman  
65 et al., 2007). Paddy fields, as major anthropogenic sources of CH<sub>4</sub> emissions, account  
66 for 5-19% of the global anthropogenic CH<sub>4</sub> budget (Smith et al., 2007). Rice is the  
67 major cereal crop for more than half of the world's population. FAO (2009) estimated  
68 that rice production must increase by 40% by the end of 2030 to meet the rising  
69 demand from the ever-increasing population. This increase may require a higher  
70 application of nitrogenous fertilizers to paddy fields, which can subsequently lead to  
71 increased emissions of CH<sub>4</sub> and N<sub>2</sub>O to the atmosphere (Kim *et al.*, 2014; Roy *et al.*,  
72 2014; Haque *et al.*, 2015).

73 To achieve both sustainable rice production and GHG reduction, various  
74 strategies of agricultural management are being developed, e.g. water management

75 (Ma et al., 2013), cultivation methods (Liu et al., 2013), fertilization schemes (Cai et  
76 al., 1997), and the development of new rice varieties (Ma et al., 2012). The  
77 application of different materials such as biochar (Zhang et al., 2010), steel slag  
78 (Wang et al., 2012), and rice straw ash (Müller-Stöver et al., 2012) are typical methods  
79 of amending agricultural soil quality and improving the production of rice paddies.  
80 However, currently most paddy fields in China are submitted to rice straw application  
81 after rice harvest, and the rice straw is burnt in situ in the paddy fields. Rice produces  
82 a large amount of agricultural residues, which is either removed from the field, burned  
83 in situ, piled or spread in the field, incorporated into the soil, or used as mulch for the  
84 following crop (Vibol and Towprayoon, 2010). Recent studies have examined the  
85 effect of straw application on GHG emission from paddy fields but have reported  
86 varying results. Several studies reported an increase in CH<sub>4</sub> emission following the  
87 addition of straw (Xu et al., 2000; Zhang et al., 2011; Yuan et al., 2014), whereas  
88 other study reported a drop in emission (Zhang et al., 2013). Also, previous studies  
89 have shown that straw application could lead to either a decrease (Zou et al, 2005) or  
90 increase (Liang et al., 2007; Zhang et al., 2013) in N<sub>2</sub>O emissions. These contrasting  
91 results may in part be attributed to differences in management practices (e.g. type and  
92 amount of fertilizer, water management, or mode of cultivation). The application of  
93 rice straw has been recommended for increasing carbon sequestration by soils (Pan et  
94 al., 2004), as a source of nutrients, and as a method for sustainable crop production  
95 (Qiu et al., 2012; Yao et al., 2015). Despite of this, most of the literature has shown so  
96 far that its effectiveness in mitigating emissions of GHGs from paddy fields is weak.

97 Usually the increases in methane emission more than counteract the occasional  
98 decrease in nitrous oxide emission (Xu et al., 2000; Zhang et al., 2011; Xia et al.,  
99 2014; Yuan et al., 2014). Conversely, other studies have also observed a reduction of  
100 emissions of GHGs related with rice straw application (Luo et al., 2010; Zhang et al.,  
101 2013) all together suggesting that the effects of straw application to paddy fields  
102 strongly depend of environmental factors such as management (flooding  
103 management) or climatic conditions. Because 90% of the paddy fields in China are in  
104 subtropical regions, such as Fujian, Jiangxi, and Hunan Provinces, the development of  
105 valid and reliable methods for reducing CH<sub>4</sub> and N<sub>2</sub>O emissions in Chinese  
106 subtropical paddy fields is of paramount importance.

107 CH<sub>4</sub> and N<sub>2</sub>O emissions are related with several soil properties, such as mainly  
108 soil temperature (Luo et al., 2013; Wang et al., 2015), soil pH (Chauhan et al., 2015;  
109 Wang et al., 2015), soil Eh (Hou et al., 2000; Johnson-Beebout et al., 2009), and soil  
110 salinity (Chauhan et al., 2015; Livesley and Andrusiak, 2012). However, the  
111 relationships between soil properties and CH<sub>4</sub> and N<sub>2</sub>O emissions are unclear and  
112 seem to depend on the ecosystem types. If the application of rice straw can effectively  
113 increase rice yield and reduce GHG emissions, the potential for widespread adoption  
114 in the paddy fields of south eastern China will be large due to the substantial amount  
115 of straw produced annually. This study: (1) examined the effects of straw application  
116 on soil properties and CH<sub>4</sub> and N<sub>2</sub>O emissions, and (2) investigated the temporal  
117 relationships between soil properties and GHG emissions.

118

119 **2. Methods**

120 *2.1 Study site*

121 This study was conducted at the Wufeng Agronomy Field of the Fujian Academy of  
122 Agricultural Sciences in Fujian Province, southeastern China (25°59'44.12"N,  
123 119°38'35.50"E) (Figure 1). Fujian province has two main crop seasons for paddy  
124 farming, an early rainy season in which rice paddies are cultivated in March or April  
125 and harvested in June or July, and a later dry season in which paddies are cultivated in  
126 July or August and harvested in November or December. We conducted field  
127 experiments during both the early rainy season (5 April - 25 July) and the late dry  
128 season (1 August - 6 November) in 2012 on successive crops in the same paddy field.  
129 The soil of the paddy field was poorly drained, and the proportions of sand, silt, and  
130 clay in the top 15 cm of the soil were 28, 60, and 12%, respectively. Other soil  
131 properties in this soil layer at the onset of the experiment were: bulk density, 1.1 g  
132 cm<sup>-3</sup>; pH (1:5 with H<sub>2</sub>O), 6.5; organic-carbon content, 18.1 g kg<sup>-1</sup>; total nitrogen (N)  
133 content, 1.2 g kg<sup>-1</sup>; and total phosphorus (P) content, 1.1 g kg<sup>-1</sup> (Wang et al., 2013 and  
134 2015a). The water level was maintained at 5-7 cm above the soil surface until the rice  
135 tiller stage, and at the late period of the rice tiller, the water was drained for about  
136 one week. After this period, the water was intermittently irrigated, until the final  
137 drainage two weeks before the rice was harvested. Irrigation water was released  
138 through an outlet.

139 The design consisted of triplicated control plots (without rice straw application)  
140 and triplicated treated plots (3.3 Mg rice straw ha<sup>-1</sup>) where rice straw was applied



141 before the transplantation of both crops. The experimental field had three independent  
142 blocks for triplicate replication. Each block contained two treatment plots (24 m<sup>2</sup>  
143 each) arranged in a randomised block design. The rice variety was Hesheng 10, and  
144 the spacing among the individual rice was 14 cm x 28 cm. The straw amendment was  
145 applied in different plots for the two crops to avoid any carry-over from the early to  
146 the late crop, although field observation indicated that almost none of the added straw  
147 remained in the soil after the harvest of the early crop. Immediately after paddy soil  
148 was plowed, we spread the rice straw by hand.

149 The field was plowed to a depth of 15 cm with a moldboard plow and leveled  
150 two days before rice transplantation. Mineral fertilizers were applied at three times  
151 with different nutrient loadings using combinations of complete (16:16:16%  
152 N:P<sub>2</sub>O<sub>5</sub>:K<sub>2</sub>O) and urea (46% N) fertilizers. The basal fertilizer was applied one day  
153 before transplanting at rates of 42 kg N ha<sup>-1</sup>, 40 kg P<sub>2</sub>O<sub>5</sub> ha<sup>-1</sup>, and 40 kg K<sub>2</sub>O ha<sup>-1</sup> and  
154 was incorporated mechanically into the top 15 cm of the soil. The tillering fertilizer  
155 applied at the tillering initiation stage (one week after transplanting) was broadcasted  
156 at rates of 35 kg N ha<sup>-1</sup>, 20 kg P<sub>2</sub>O<sub>5</sub> ha<sup>-1</sup>, and 20 kg K<sub>2</sub>O ha<sup>-1</sup>. The final topdressing  
157 fertilizer applied eight weeks after transplantation at the start of panicle formation was  
158 broadcasted at rates of 18 kg N ha<sup>-1</sup>, 10 kg P<sub>2</sub>O<sub>5</sub> ha<sup>-1</sup>, and 10 kg K<sub>2</sub>O ha<sup>-1</sup>. Both crops  
159 received the same management. The early and late crops were harvested on 25 July  
160 and 6 November, respectively.

161

## 162 *2.2 Measurement of CH<sub>4</sub> and N<sub>2</sub>O emissions*

163 CH<sub>4</sub> and N<sub>2</sub>O fluxes were measured during growth using PVC static closed squared  
164 chambers as described by Datta et al. (2013). The chambers consisted of two parts, a

165 permanently installed bottom collar (30 cm length, 30 cm width, 10 cm height) and a  
166 removable, transparent upper compartment (30 cm length, 30 cm width, 100 cm  
167 height). Three chambers were deployed in each plot, each covering three rice hills.  
168 Each chamber had two battery-operated fans to mix the air inside the chamber  
169 headspace, a thermometer to monitor temperature changes during sampling, and a gas  
170 sampling port with a neoprene septum at the top. A wooden boardwalk was built for  
171 accessing the plots to minimize soil disturbance during sampling.

172 Gas fluxes were measured in all chambers at intervals of 1-2 weeks. Gas samples  
173 were collected from the chamber headspace by a 100-ml plastic syringe equipped with  
174 a 3-way stopcock 0, 15, and 30 min after chamber deployment. Samples were  
175 collected twice a day to obtain reliable estimates of the daily mean CH<sub>4</sub> and N<sub>2</sub>O  
176 fluxes. The samples were taken between 9:00 and 11:00 am (Wang, 2001; Wang et al.,  
177 2015b). The samples were immediately transferred to 100-ml air-evacuated  
178 aluminum-foil bags (Delin Gas Packaging Co., Ltd., Dalian, China) sealed with  
179 butyl-rubber septa and were transported to the laboratory for analysis

180

### 181 2.3 Determination of CH<sub>4</sub> and N<sub>2</sub>O concentrations

182 CH<sub>4</sub> and N<sub>2</sub>O concentrations in the headspace air samples were determined with a  
183 GC-2014 gas chromatograph with a Porapak Q stainless steel column (2 m length, 4  
184 mm OD, 80/100 mesh) (Shimadzu, Kyoto, Japan). CH<sub>4</sub> and N<sub>2</sub>O were detected with a  
185 flame ionization detector (FID) and an electron capture detector (ECD), respectively.

186 The operating temperatures of the column, injector, and detector were adjusted to 70,

187 200, and 200 °C, respectively, for the determination of CH<sub>4</sub> concentrations and to 70,  
188 200, and 320 °C, respectively, for the determination of N<sub>2</sub>O concentrations. Helium  
189 (99.999% purity) was used as a carrier gas (30 ml min<sup>-1</sup>), and a make-up gas (95%  
190 argon and 5% CH<sub>4</sub>) was used for the electron capture detector. The gas  
191 chromatograph was calibrated before and after each set of measurements using 1.01,  
192 7.99, and 50.5 µl CH<sub>4</sub> l<sup>-1</sup> in helium and 0.2, 0.6, and 1.0 µl N<sub>2</sub>O l<sup>-1</sup> in helium  
193 (CRM/RM information center of China) as primary standards.

194

#### 195 2.4 Calculation of CH<sub>4</sub> and N<sub>2</sub>O fluxes

196 CH<sub>4</sub> and N<sub>2</sub>O fluxes from the paddy field were expressed as the rate of change in the  
197 mass of CH<sub>4</sub> and N<sub>2</sub>O per unit surface area per unit time. CH<sub>4</sub> and N<sub>2</sub>O fluxes were  
198 calculated by (Ali et al., 2008):

199

$$200 \quad F = \frac{M}{V} \cdot \frac{dc}{dt} \cdot H \cdot \left( \frac{273}{273 + T} \right)$$

201 where  $F$  is the CH<sub>4</sub> or N<sub>2</sub>O flux (mg CH<sub>4</sub> m<sup>-2</sup> h<sup>-1</sup> or µg N<sub>2</sub>O m<sup>-2</sup> h<sup>-1</sup>),  $M$  is the molar  
202 mass of the gas (16 for CH<sub>4</sub> and 44 for N<sub>2</sub>O),  $V$  is the standard molar volume of air  
203 (22.4 l mol<sup>-1</sup>),  $dc/dt$  is the change in headspace CH<sub>4</sub> or N<sub>2</sub>O concentration with time  
204 (µmol mol<sup>-1</sup> h<sup>-1</sup>),  $H$  is the height of the chamber above the water surface (m), and  $T$  is  
205 the air temperature inside the chamber (°C).

206

#### 207 2.5 Global warming potential (GWP)

208 To estimate GWP, CO<sub>2</sub> is typically taken as the reference gas, and a change in the  
209 emission of CH<sub>4</sub> or N<sub>2</sub>O is converted into “CO<sub>2</sub>-equivalents” (Hou et al., 2012). The

210 GWP for CH<sub>4</sub> is 34 (based on a 100-year time horizon and a GWP for CO<sub>2</sub> of 1), and  
211 the GWP for N<sub>2</sub>O is 298 (Myhre et al., 2013). Cumulative CH<sub>4</sub> emission and N<sub>2</sub>O  
212 emission were calculated from average measurements through time. The GWP of the  
213 combined emission of CH<sub>4</sub> and N<sub>2</sub>O was calculated using the equation (Ahmad et al.,  
214 2009):

215

$$216 \quad \text{GWP} = \text{cumulative CH}_4 \text{ emission} \times 34 + \text{cumulative N}_2\text{O emission} \times 298$$

217

## 218 *2.6 Measurement of soil properties*

219 Soil temperature (°C), pH, salinity (mS cm<sup>-1</sup>), and redox potential (Eh, mV) in each plot  
220 were measured *in situ* in triplicate from the upper 0-15 cm of soil in each sampling day.  
221 Eh, pH, and temperature were measured with an Eh/pH/temperature meter with  
222 internal reference electrode (IQ Scientific Instruments, Carlsbad, CA, USA), and soil  
223 salinity was measured with a 2265FS EC Meter (Spectrum Technologies Inc., Aurora,  
224 IL, USA).

225

## 226 *2.7 Statistical analysis*

227 All statistical analyses were performed using SPSS Statistics 18.0 (SPSS Inc.,  
228 Chicago, USA). Differences in CH<sub>4</sub> and N<sub>2</sub>O fluxes and soil properties between  
229 sampling dates and plots with and without straw application were determined by  
230 two-way repeated-measures ANOVAs. The relationships between CH<sub>4</sub> and N<sub>2</sub>O flux  
231 and the soil properties in the treated and control plots were determined by Pearson  
232 correlation analysis.

233 **3. Results**

234 *3.1 CH<sub>4</sub> emissions from the crops*

235 The emission of CH<sub>4</sub> in the early crop varied significantly across sampling dates and  
236 for the interactions between treatment and sampling date ( $P < 0.01$ , Table S1) but not  
237 between the treatments ( $P > 0.05$ , Table S1). Figure 2 shows the seasonal variations of  
238 CH<sub>4</sub> emissions in the crops during the growing period. Fluxes were relatively low  
239 (0.02-0.36 mg m<sup>-2</sup> h<sup>-1</sup>) during the initial growth period of the early crop (6-20 April).  
240 The fluxes, however, increased to peaks of 0.53 and 1.79 mg m<sup>-2</sup> h<sup>-1</sup> on 5 May in the  
241 treated and control plots, respectively, as the duration of flooding increased. The  
242 fluxes decreased considerably to  $< 0.05$  mg m<sup>-2</sup> h<sup>-1</sup> on 19 May during the final tillering  
243 stage following drainage of the field and then increased gradually to second peaks of  
244 1.00 and 0.77 mg m<sup>-2</sup> h<sup>-1</sup> on 28 June in the treated and control plots, respectively,  
245 following re-flooding one week after drainage. CH<sub>4</sub> emissions then decreased steadily  
246 until the rice was harvested when the field was completely drained.

247 CH<sub>4</sub> emissions from the late crop differed significantly across treatments and  
248 sampling dates for and the interactions between treatment and sampling date ( $P < 0.01$ ,  
249 Table S1). The fluxes were significantly higher in the treated than the control plots  
250 ( $P < 0.05$ , Figure 2) during the regreening and tillering stages between 6 August and 3  
251 September. Only one CH<sub>4</sub> flux peak was observed, on 13 August, for both the treated  
252 and control plots (94.72 and 16.90 mg m<sup>-2</sup> h<sup>-1</sup>, respectively), in contrast to the double  
253 peaks in the early crop. Even though the paddy field was re-flooded on 17 September,  
254 the CH<sub>4</sub> flux remained low ( $< 1.00$  mg m<sup>-2</sup> h<sup>-1</sup>) during the later period of growth until

255 harvesting in November. Mean CH<sub>4</sub> flux was significantly higher for the treated than  
256 the control plots in the late crop ( $P<0.05$ ) but not the early crop ( $P<0.05$ ) (Data not  
257 shown).

258

### 259 *3.2 Nitrous oxide emissions from the crops*

260 N<sub>2</sub>O fluxes in the early crop varied significantly across sampling dates and for the  
261 interactions between treatment and sampling date ( $P<0.01$ , Table S1) but not between  
262 the treatments ( $P>0.05$ , Table S1). Figure 3 shows the seasonal variations of N<sub>2</sub>O flux  
263 in the crops during the growing period. The temporal pattern of N<sub>2</sub>O flux was not  
264 consistent between the treated and control plots. The control plots in the early crop  
265 had only one emission peak (5 May, 133.03  $\mu\text{g m}^{-2} \text{h}^{-1}$ ), but the treated plots in the  
266 late crop had two peaks, on 6 April during the early stage after transplanting (57.58  $\mu\text{g}$   
267  $\text{m}^{-2} \text{h}^{-1}$ ) and on 3 June after the drainage and re-flooding of the field at the late tillering  
268 stage (91.80  $\mu\text{g m}^{-2} \text{h}^{-1}$ ).

269 N<sub>2</sub>O fluxes in the late crop differed significantly among sampling dates and for  
270 the interactions between treatment and sampling date ( $P<0.01$ , Table S1, Figure 3) but  
271 not between the treatments ( $P>0.05$ , Table S1, Figure 3). The control plots had two  
272 flux maxima, the first (63.73  $\mu\text{g m}^{-2} \text{h}^{-1}$ ) on 17 September following the re-flooding of  
273 the paddy field, and the second (52.12  $\mu\text{g m}^{-2} \text{h}^{-1}$ ) on 29 October when the field was  
274 drained during the late maturity stage. The treated plots had only one N<sub>2</sub>O flux peak,  
275 on 6 August (70.02  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). Minimum N<sub>2</sub>O flux occurred on 3 September in the  
276 control plots (-55.67  $\mu\text{g m}^{-2} \text{h}^{-1}$ ) but was earlier (13 August) in the treated plots

277 (-28.49  $\mu\text{g m}^{-2} \text{h}^{-1}$ ). Mean  $\text{N}_2\text{O}$  emissions in the early crop were significantly lower in  
278 the treated than the control plots ( $P<0.05$ , Figure 3). In contrast, mean  $\text{N}_2\text{O}$  emissions  
279 in the late crop were significantly higher in the treated than the control plots ( $P<0.05$ ,  
280 Figure 3).

281

### 282 *3.3 Variations in soil properties during the experimental period*

283 Temperatures in the top 15 cm of the soil in the early crop differed significantly  
284 among sampling dates and for the interactions between treatment and sampling date  
285 ( $P<0.01$ ) but not between the treatments ( $P>0.05$ , Table S2). Soil temperature in the  
286 late crop differed significantly among sampling dates ( $P<0.01$ ) but not for the  
287 interactions between treatment and sampling date or between the treatments ( $P>0.05$ ,  
288 Table S2). Soil temperature generally increased from 19 °C at the beginning of the  
289 experimental period to a peak of 32 °C on 9 July and then decreased gradually  
290 throughout the growing period of the late crop (Figure 4).

291 Soil pH and salinity in the early crop differed significantly among sampling dates  
292 ( $P<0.01$ , Table S2, Figure 4) but not for the interactions between treatment and  
293 sampling date or between the treatments ( $P>0.05$ , Table S2, Figure 4). Soil pH in the  
294 late crop differed significantly among sampling dates and for the interactions between  
295 treatment and sampling date ( $P<0.01$ , Table S2, Figure 4) but not between the  
296 treatments ( $P>0.05$ , Table S2, Figure 4). Soil Eh in both crops differed significantly  
297 among sampling dates and for the interactions between treatment and sampling date  
298 ( $P<0.01$ , Table S2, Figure 4) but not between the treatments ( $P>0.05$ , Table S2, Figure

299 4).

300 Mean soil temperature, pH, salinity, and Eh in the early crop did not differ  
301 significantly between the treatments ( $P>0.05$ , Table 1). In the late crop, soil salinity  
302 was significantly higher and soil Eh was significantly lower in the treated than the  
303 control plots ( $P<0.05$ , Table 1).

304

### 305 *3.4 Relationships between CH<sub>4</sub> and N<sub>2</sub>O emissions and soil properties*

306 Table 2 shows the Pearson correlation coefficients between CH<sub>4</sub> and N<sub>2</sub>O emissions  
307 and soil properties during the growing periods of the two crops of rice. CH<sub>4</sub> emission  
308 was significantly correlated negatively with soil pH ( $r = -0.32$ ,  $P<0.05$ ) in the treated  
309 plots of the early crop and positively with soil temperature ( $r = 0.58-0.64$ ) and salinity  
310 ( $r = 0.67-0.72$ ) in both treatments in the late crop ( $P<0.01$ ).

311 N<sub>2</sub>O emission in the control plots of the early crop was correlated negatively  
312 with soil temperature and Eh ( $r = -0.37$ ,  $P<0.05$ ) and positively with soil pH ( $r = 0.34$ ,  
313  $P<0.05$ ). N<sub>2</sub>O emission in the treated plots was significantly ( $P<0.05$ ) correlated  
314 negatively with soil pH ( $r = -0.40$ ) and salinity ( $r = -0.42$ ) and positively with soil Eh  
315 ( $r = 0.41$ ). N<sub>2</sub>O emission in the control plots of the late crop was significantly ( $P<0.05$ )  
316 correlated negatively with soil temperature ( $r = -0.26$ ), pH ( $r = -0.53$ ), and salinity ( $r$   
317  $= -0.31$ ) and positively with soil Eh ( $r = 0.54$ ).

318

### 319 *3.5 Global warming potential*

320 The influence of paddy fields on the radiative forcing of the atmosphere and hence



321 climate change can be assessed by determining the GWP from the  
322 biosphere-atmosphere exchange of various GHGs. The GWP contributed by CH<sub>4</sub>  
323 emissions in the early crop did not differ significantly between the treatments ( $P>0.05$ ,  
324 Table 3). The GWP due to N<sub>2</sub>O emissions and the combined CH<sub>4</sub> and N<sub>2</sub>O emissions,  
325 however, was significantly lower in the treated than the control plots ( $P<0.05$ , Table 3).  
326 The GWPs in the late crop due to CH<sub>4</sub> or N<sub>2</sub>O emissions alone and to the combined  
327 emissions were all significantly higher in the treated than the control plots ( $P<0.05$ ,  
328 Table 3).

329

### 330 3.6. Rice yield

331 The rice yield was  $4.68 \pm 0.17$  and  $5.13 \pm 0.23$  (Mg ha<sup>-1</sup>) ( $P<0.001$ ) in control and  
332 straw treated plots in the early rice, respectively, and  $6.27 \pm 0.15$  and  $6.56 \pm 0.28$  (Mg  
333 ha<sup>-1</sup>) ( $P<0.001$ ) in the control and straw treated plots in the late rice, respectively  
334 (Figure 5).

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343 **4. Discussion**

344 *4.1 Seasonal variations of CH<sub>4</sub> and N<sub>2</sub>O emissions and relationships with soil*  
345 *properties*

346 The rate of CH<sub>4</sub> emission in the early crop was relatively low at the initial growing  
347 stage but then increased quickly to the first emission peak on 5<sup>th</sup> May during the  
348 tillering period. This peak was due predominantly to the development of anaerobic  
349 soil conditions and to the rapid growth of the rice plants that facilitated the  
350 plant-mediated transport of CH<sub>4</sub>. CH<sub>4</sub> emissions decreased substantially after this  
351 peak due to the drainage of the paddy field and hence to an increase in the soil redox  
352 potential before rising to another peak on 28 June during the heading and flowering  
353 period. This second peak could mostly be attributed to the large supply of litter and  
354 root exudates during the later stage of rice growth. This addition of carbon sources  
355 will increase the availability of substrates for methanogens, which will then increase  
356 CH<sub>4</sub> production and eventually emission to the atmosphere (Kimura et al., 2004;  
357 Gaihre et al., 2011). Moreover, plant-mediated transport of CH<sub>4</sub> is particularly  
358 efficient at this stage of plant growth because of the well-developed aerenchymatous  
359 system (Wang et al., 2014b). Eh control methane production, but methane emission  
360 depends of other several processes such as oxidation and transportation that depend of  
361 several other factors. In other studies in this area not significant correlations were  
362 either found between methane emission and Eh (Tong et al., 2010; Wang et al., 2015).

363 CH<sub>4</sub> emissions in both crops peaked in the tillering period and then decreased  
364 gradually due to the complete drainage of the paddy field, which is a necessary

365 management practice during the latter part of tillering (Figure 2) because the drainage  
366 was to use finish the tiller process of rice (Zhu, 2010). This temporal variability was  
367 consistent with the patterns observed in other studies of CH<sub>4</sub> flux in paddy fields (Jia  
368 et al. 2001; Ali et al., 2013; Kim et al., 2013). Towprayoon et al. (2005) similarly  
369 found a decrease in CH<sub>4</sub> emission as a result of midseason and multiple drainages,  
370 which could be attributed to the increasingly oxic conditions in the sediments that  
371 suppress methanogenesis and thus CH<sub>4</sub> emissions (Tsuruta, 2002; Singh et al., 2003).  
372 The major difference in flux pattern between the early and late crops was the absence  
373 of a second CH<sub>4</sub> peak in the late crop. Despite the re-flooding of the paddy field after  
374 drainage at the final tillering stage, CH<sub>4</sub> emission did not further increase in the late  
375 crop due to the relatively low temperatures during this period. The role of temperature  
376 as a limiting factor of CH<sub>4</sub> emission has been discussed in Gaihre et al. (2013). In  
377 contrast, CH<sub>4</sub> emissions in the early crop increased to a second peak on 28 June  
378 subsequent to re-flooding, because the combination of high temperatures and  
379 anaerobic conditions strongly favor methanogenesis.

380 N<sub>2</sub>O emissions in the treated plots of the early crop had two seasonal peaks, with  
381 the first in early April and the second in early June. Both maxima occurred when the  
382 paddy field was re-flooded after a period of complete drainage. Our results suggest  
383 that the alternate wetting and drying of the field plays a crucial role in facilitating the  
384 production and emission of N<sub>2</sub>O. This premise supports the findings of greenhouse  
385 experiments by Johnson-Beebout et al. (2009) that alternate wetting and drying  
386 increased N<sub>2</sub>O emissions from paddy soils relative to a continuously flooded

387 treatment. The timing of the minimum N<sub>2</sub>O emission in our study differed between  
388 treatments, but the lowest N<sub>2</sub>O emission, or the highest rate of N<sub>2</sub>O uptake, occurred  
389 consistently after the field was flooded for approximately one month (May and June  
390 for the treated and control plots, respectively). Prolonged flooding promotes the  
391 development of strong anaerobic conditions in soils, which promotes the reduction of  
392 any N<sub>2</sub>O produced from the paddy fields to N<sub>2</sub> (Ussiri and Lal, 2013). The treated  
393 plots of the late crop had only one peak, in August, probably because the rapid  
394 decomposition of the straw under the high temperatures provided a large supply of  
395 labile substrates for microbes to produce N<sub>2</sub>O (Zhang et al., 2013).

396 CH<sub>4</sub> emissions in the early crop were significantly negatively correlated with soil  
397 pH only in the treated plots, suggesting that the methanogens may have been better  
398 adapted to the acidic paddy soil, in contrast to the findings by Valentine et al. (1994)  
399 that the potential for CH<sub>4</sub> production decreased significantly as pH decreased from 7  
400 to 5.5 in peat in northern Canadian fens. Better soil conditions for CH<sub>4</sub>  
401 production-emissions could be generated in conditions of excess of soil organic  
402 carbon such as under straw application. Under favorable conditions methane  
403 production shoots up when soil organic carbon is suddenly enhanced by straw  
404 application (Ye et al., 2015). CH<sub>4</sub> emission in the late crop was significantly positively  
405 correlated with soil temperature, which may have been due to the  
406 temperature-enhanced microbial CH<sub>4</sub> production. This positive effect of temperature  
407 on CH<sub>4</sub> emission has also been seen in temperate spruce forests in Germany, tropical  
408 rain forests in Australia, and ungrazed semi-arid steppes in China (Luo et al., 2013).

409 Furthermore, CH<sub>4</sub> emission was significantly positively correlated with soil salinity in  
410 both treatments. N<sub>2</sub>O emissions in the control plots of both our crops were  
411 consistently negatively correlated with soil temperature, which implied that  
412 temperature was not a limiting factor for N<sub>2</sub>O emission.

413

#### 414 *4.2 Effects of straw application on soil properties*

415 Soil Eh generally decreased in response to rice straw application, similar to the  
416 findings in another study (Gaihre et al., 2013), which could be attributed to a number  
417 of reasons. Firstly, the decomposition of rice straw will increase the supply of  
418 electrons for reduction reactions, thereby lowering soil Eh (Gao et al., 2004;  
419 Minamikawa and Sakai, 2006). Secondly, rice straw has a high ability to absorb  
420 moisture and hence to maintain a more anaerobic soil environment. Our application of  
421 rice straw also led to increases in both soil salinity and pH. Rice straw contains  
422 numerous elements essential for plant growth, including nitrogen, phosphorus,  
423 potassium, calcium, and sodium (Gaihre et al., 2013; Zhang et al., 2013). Many of  
424 these cationic nutrients will be returned to the soil solution following the  
425 decomposition of the straw, which in turn will increase soil salinity and conductivity.

426

#### 427 *4.3 Effects of straw application on CH<sub>4</sub> and N<sub>2</sub>O emissions*

428 Mean CH<sub>4</sub> emission in the early crop did not differ significantly between the  
429 treatments. The early crop was grown in the rainy season when temperatures were  
430 lower than for the late crop, so the decomposition of the straw was slower, which

431 supplied less labile carbon for CH<sub>4</sub> production. The temperature increased during the  
432 early stages of rice growth, so the rate of straw decomposition and the carbon input to  
433 the soil also increased, but the end of this stage coincided with soil drainage and the  
434 consequent aerobic soil conditions that are not suitable for methane production. These  
435 environmental conditions unfavorable for methane production in the early crop were  
436 likely responsible for the lack of differences in cumulative CH<sub>4</sub> emission between the  
437 treatments.

438 N<sub>2</sub>O emissions decreased following the addition of the rice straw. Straw  
439 application will inevitably lead to an increase in the amount of soil organic carbon,  
440 mostly toward the end of the early stage of rice growth as temperatures increase,  
441 which subsequently leads to an increased demand for N by soil microbes. The  
442 increases in N content of the microbe biomass as the soil becomes more N-limited  
443 would lead to an overall decrease in N<sub>2</sub>O emissions from the soil. Previous studies  
444 have also found that N-limitation decreased N<sub>2</sub>O emission in paddy fields (Zhang et  
445 al., 2010).

446 In contrast, mean CH<sub>4</sub> emissions during the late crop were significantly higher in  
447 the treated than the control plots. The rice straw in this crop was applied in summer at  
448 higher temperatures. The soil organic-carbon content (18.1 g kg<sup>-1</sup>) at our study site  
449 was lower than that reported for paddy fields in Tsuruoka, Japan (29.0 g kg<sup>-1</sup>) (Wang  
450 et al., 2012; Itoh et al., 2011). CH<sub>4</sub> production in our study site was thus probably  
451 limited by the availability of labile carbon substrates, which is consistent with the  
452 positive response of CH<sub>4</sub> production to carbon addition in wetland soils in the same

453 study area (Wang et al., 2008). The roots of the rice cultivar used in the present study  
454 may not exude much labile organic carbon for methanogenesis in the rhizosphere.  
455 Sigren et al. (1997) have reported significant variations in the acetate concentrations  
456 in root exudates and seasonal CH<sub>4</sub> emissions between two rice cultivars in the same  
457 field.

458 Mean N<sub>2</sub>O emission in the late crop was significantly higher in the treated than  
459 the control plots. Rice paddies have a relatively high N demand and hence incorporate  
460 a large amount of N in their biomass during growth (Zhao, 2012). One possibility to  
461 explain this higher N<sub>2</sub>O emission is that the rapid decomposition of the straw from the  
462 early crop during the growth of the late rice crop period would provide a substantial  
463 input of N to the soils, which would provide substrates necessary for N<sub>2</sub>O production  
464 and thus higher N<sub>2</sub>O emissions. This additional supply of N is particularly important  
465 in governing N<sub>2</sub>O dynamics in this region of southeastern China, which is N-limited.  
466 Wang et al. (2012) found that total soil N concentrations actually decreased in  
467 cultivated rice-paddy soils despite the addition of 95 kg N ha<sup>-1</sup> during the growth  
468 period, which suggests a very rapid rate of N uptake by the crop. However, to confirm  
469 or not the possibility that higher N release after straw application is underlying higher  
470 N<sub>2</sub>O emissions will need further experiments.

471 The higher N<sub>2</sub>O emissions observed in the early crop than in the late can be  
472 related to the N fertilizer addition in equal amounts during the two growing stages.  
473 The addition of the same amount of fertilizer to two crops which have quite different  
474 length of growing season probably means that the shorter crop was over-fertilized,

475 which might partially explain the overall higher N<sub>2</sub>O flux in the early crop than in the  
476 late. Applying the same amount of N to plots with and without straw is likely to  
477 affect differently the yield in two growth periods, because straw decomposition  
478 usually causes early-season N immobilization and this would be more detrimental to  
479 plant N uptake in the short growth period (early) than in the longer (late).

480

#### 481 4.4 Potential of straw application in paddy-field management

482 The results of our study indicated that CH<sub>4</sub> emissions varied seasonally between the  
483 two crops of rice, with two seasonal peaks in the early crop but only one in the late  
484 crop, as a result of different temperatures during the two growth periods. Despite most  
485 studies have observed that straw application can increase overall GWP emissions our  
486 study of the application of straw to the subtropical paddy soils of Fujian effectively  
487 reduced N<sub>2</sub>O emissions and the overall GWP in the early crop, thus demonstrating the  
488 potential of minimizing the adverse impacts of rice agriculture on climate change  
489 when applied in adequate environmental conditions. Straw application, however,  
490 increased both mean CH<sub>4</sub> and N<sub>2</sub>O emissions in the late crop. The results showed that  
491 rice straw application increased rice yield (9.63% -12.58%) (Pan, 2014) in early and  
492 late crop period whereas its relationships with gas emissions may change depending  
493 of the time when applied. Thus the overall results suggest that in this south China rice  
494 crops the timing of the application of rice straw is thus critical for maximizing its  
495 benefits to increase crop yields and decrease GHG and GWP emissions or at least  
496 avoid increasing them.



497

498 **Acknowledgments**

499 The authors would like to thank Yongyue Ma, Linmei Ouyang, and Xianbiao Lin for  
500 their assistance with field sampling. Funding was provided by the National Science  
501 Foundation of China (31000209), Natural Science Foundation Key Programs of  
502 Fujian Province (2014R1034-3, 2014Y0054, and 2014J01119), the Spanish  
503 Government grant CGL2013-48074-P, the Catalan Government project grant SGR  
504 2014-274, and the European Research Council Synergy grant ERC-2013-SyG-610028  
505 IMBALANCE-P.

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Accepted version

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699 **Table 1** Mean soil properties  $\pm$  standard error in the treated and control plots for the  
 700 two crops of rice.  
 701

Soil property	Early crop		Late crop	
	Control	Straw	Control	Straw
Soil temperature ( $^{\circ}\text{C}$ )	26.60 $\pm$ 0.83a	27.22 $\pm$ 0.86a	26.09 $\pm$ 0.58a	26.19 $\pm$ 0.56a
Soil pH	6.16 $\pm$ 0.07a	6.31 $\pm$ 0.09a	6.15 $\pm$ 0.06a	6.37 $\pm$ 0.07a
Soil salinity ( $\text{mS cm}^{-1}$ )	0.22 $\pm$ 0.02a	0.24 $\pm$ 0.02a	0.54 $\pm$ 0.04b	0.76 $\pm$ 0.04a
Soil Eh (mV)	48.54 $\pm$ 4.52a	41.68 $\pm$ 5.22a	49.38 $\pm$ 3.41a	35.16 $\pm$ 3.15b

702 Different letters indicate significant differences between the treated and control plots ( $P < 0.05$ ).  
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704 **Table 2** Pearson correlation coefficients between CH<sub>4</sub> and N<sub>2</sub>O emissions and soil  
 705 properties in the treated and control plots for the early (N=27) and late (N=45) crops.  
 706

Gas	Crop	Treatment	Soil temperature	Soil pH	Soil salinity	Soil Eh
CH <sub>4</sub>	Early	Control	NS	NS	NS	NS
		Straw	NS	-0.320*	NS	NS
	Late	Control	0.581**	NS	0.722**	NS
		Straw	0.635**	NS	0.667**	NS
N <sub>2</sub> O	Early	Control	-0.371*	0.338*	NS	-0.372*
		Straw	NS	-0.402*	-0.423**	0.405*
	Late	Control	-0.255*	-0.528**	-0.306*	0.538**
		Straw	NS	NS	NS	NS

707 \*P<0.05; \*\*P<0.01; NS, not significant

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708 **Table 3** Cumulative mean CH<sub>4</sub> and N<sub>2</sub>O emissions  $\pm$  standard error over the study period and the overall global warming potential (GWP) in the  
 709 treated and control plots for the two crops of rice.  
 710

Crop	CH <sub>4</sub> emission				N <sub>2</sub> O emission				Combined GWP	
	Cumulative		GWP		Cumulative		GWP		(kg CO <sub>2</sub> -eq ha <sup>-1</sup> )	
	(g m <sup>-2</sup> )		(kg CO <sub>2</sub> -eq ha <sup>-1</sup> )		(mg m <sup>-2</sup> )		(kg CO <sub>2</sub> -eq ha <sup>-1</sup> )			
	Control	Straw	Control	Straw	Control	Straw	Control	Straw	Control	Straw
Early	1.13 $\pm$ 0.17	0.83 $\pm$ 0.09	283.73 $\pm$ 42.68	206.37 $\pm$ 22.38	75.92 $\pm$ 8.05a	43.42 $\pm$ 7.46b	226.25 $\pm$ 23.99a	129.39 $\pm$ 22.23b	509.98 $\pm$ 53.82a	335.76 $\pm$ 43.61b
Late	8.34 $\pm$ 1.13b	37.27 $\pm$ 4.28a	2084.36 $\pm$ 282.41b	9317.16 $\pm$ 1069.96a	17.57 $\pm$ 3.91b	36.19 $\pm$ 5.74a	52.37 $\pm$ 11.65b	107.84 $\pm$ 17.10a	2136.73 $\pm$ 287.27b	9425.00 $\pm$ 1091.86a

711 Different letters indicate significant differences between the treated and control plots ( $P < 0.05$ ).

712 **Figure Captions**

713

714 **Figure 1.** The location of the study area and sampling site (▲) in Fujian Province,  
715 southeastern China.

716 **Figure 2.** Seasonal variation of CH<sub>4</sub> fluxes in the early (A) and late (B) crops. Error bars  
717 indicate 1 standard error of the mean of triplicate measurements. \* indicate significant  
718 differences between treatments ( $P<0.05$ ). Results come from the repeated-measures ANOVAs  
719 for soil properties between the treatments and among the sampling dates.

**Figure 3.** Seasonal  
720 variation of N<sub>2</sub>O fluxes in the early (A) and late (B) crops. Error bars indicate 1 standard error  
721 of the mean of triplicate measurements. \* indicate significant differences between treatments  
722 ( $P<0.05$ ). Results come from the repeated-measures ANOVAs for soil properties between the  
723 treatments and among the sampling dates.

**Figure 4.** Temporal variation of soil properties in the early (A) and late (B) crops. Error bars  
724 indicate 1 standard error of the mean of triplicate measurements. \* indicate significant  
725 differences between treatments ( $P<0.05$ ). Results come from the repeated-measures ANOVAs  
726 for soil properties between the treatments and among the sampling dates.

**Figure 5.** Rice production (mean  $\pm$  S.E., Mg ha<sup>-1</sup>) in control and straw fertilized plots during  
728 early and late rice crops. Different letters indicate significant differences between treatments  
729 ( $P<0.05$ ).

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732 **Figure 1**

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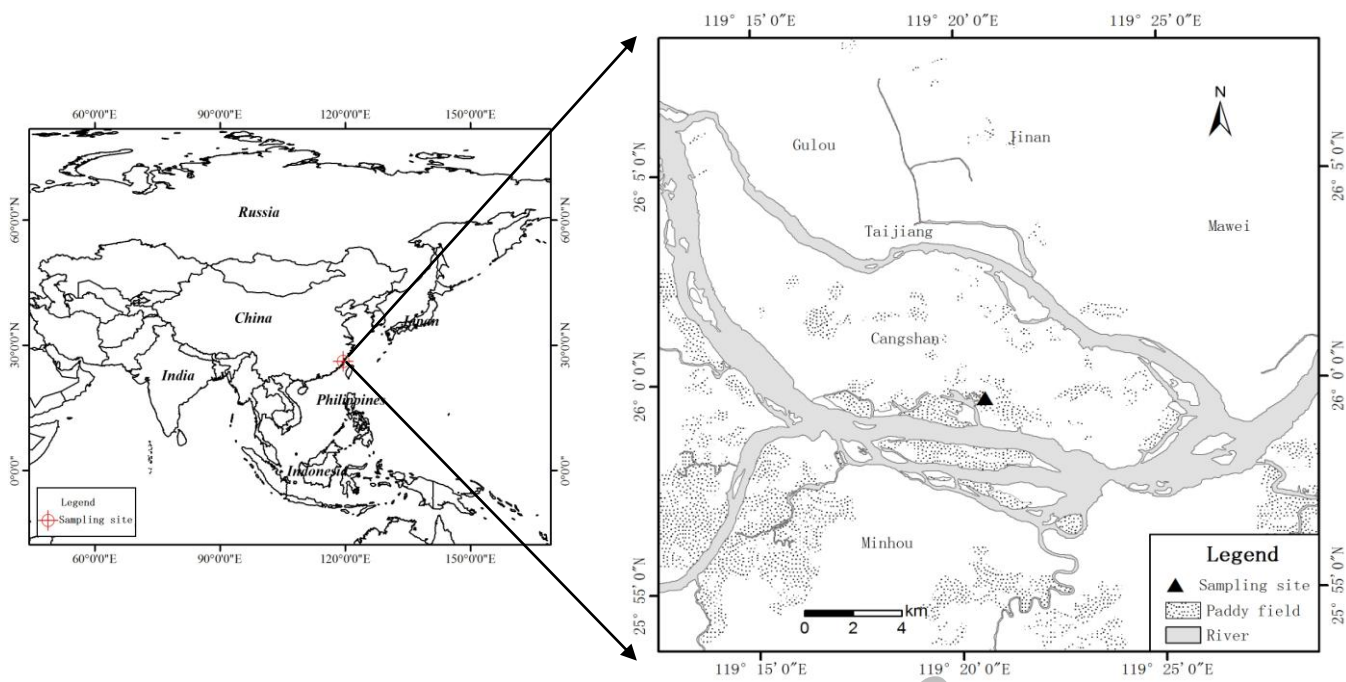
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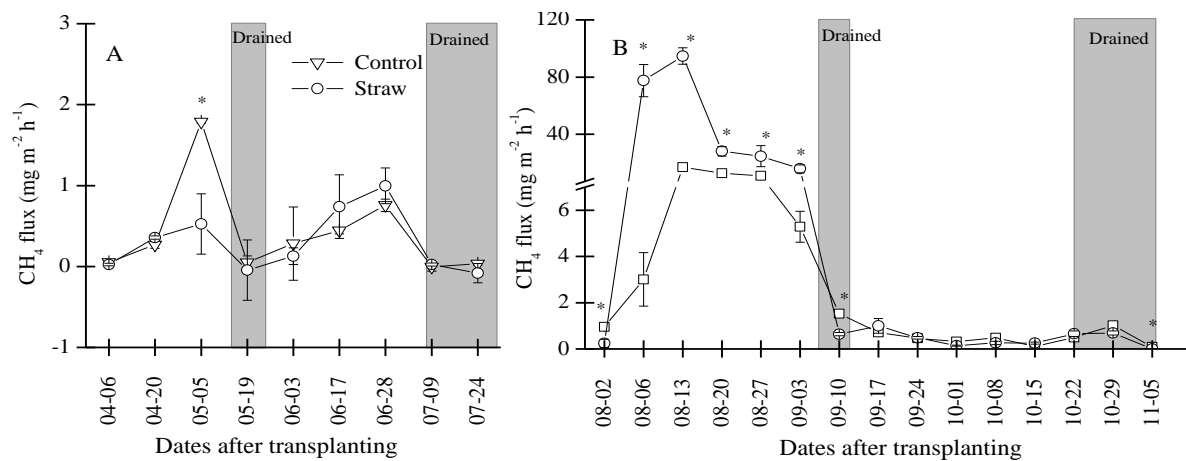
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743 **Figure 2**

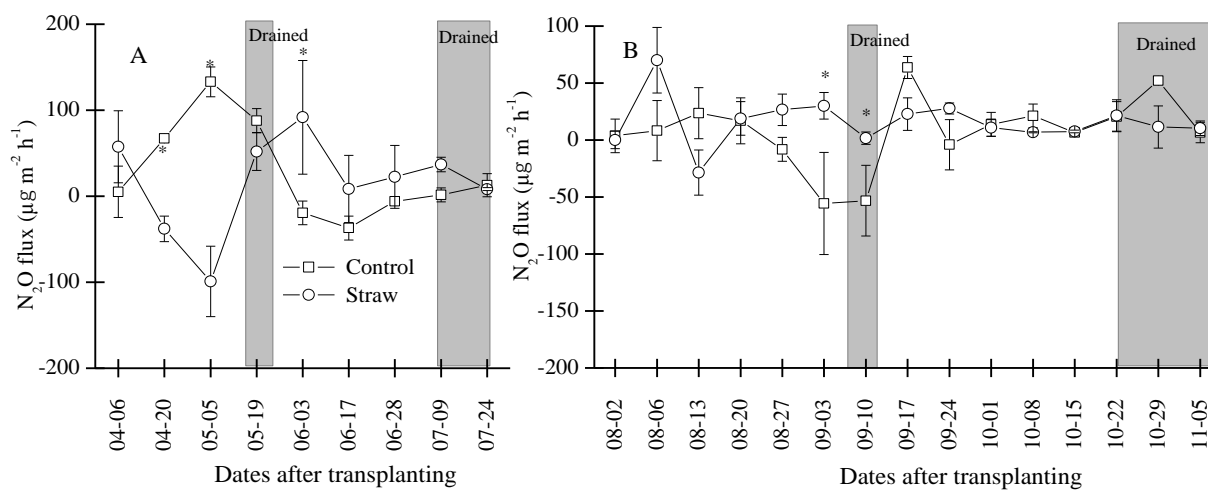


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746 **Figure 3**

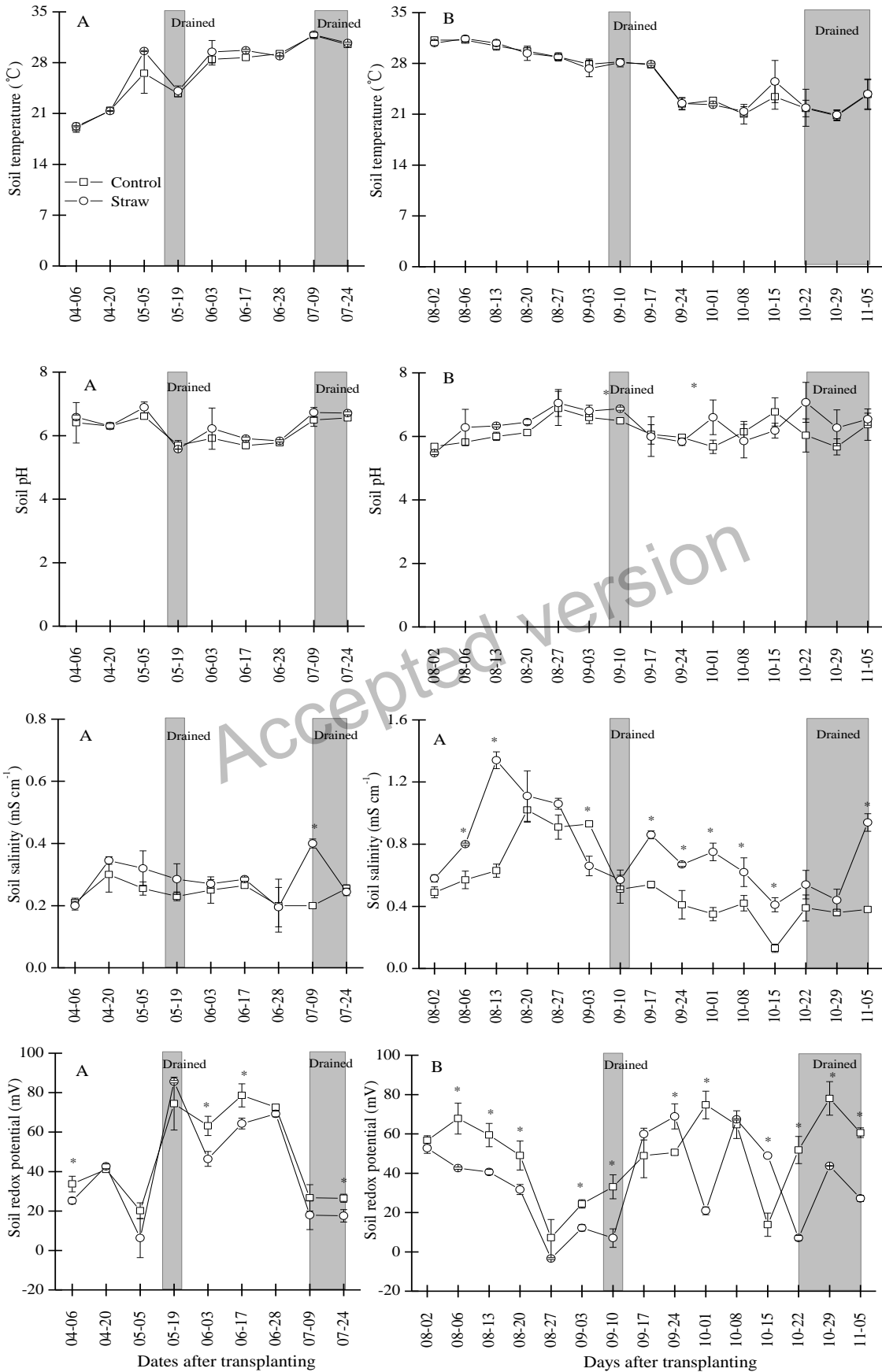


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749 **Figure 4**



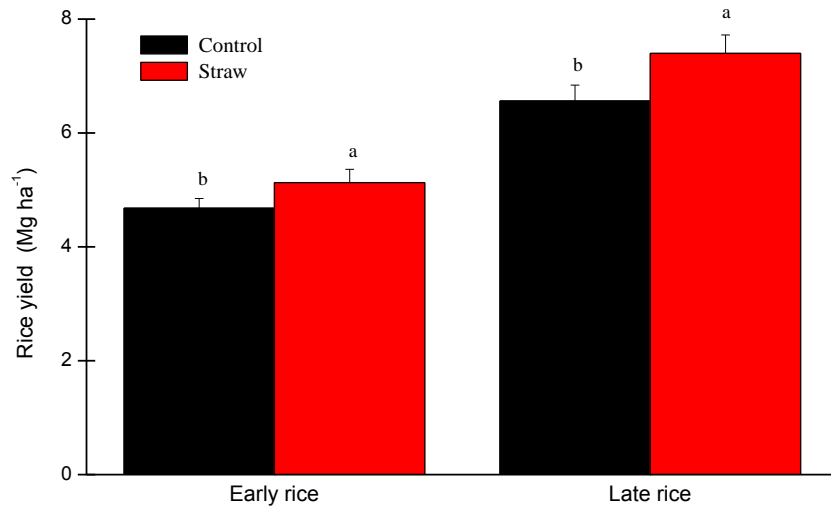
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755 Figure 5

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