# Relationships between the potential production of the greenhouse gases $CO_2$, $CH_4$ and $N_2O$ and soil concentrations of C, N and P across 26 paddy fields in southeastern China

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Abstract

Paddy fields are a major global anthropogenic source of greenhouse gases. China has the second largest area under rice cultivation, so determining the relationships between the emission of greenhouse gases and soil carbon content, nutrient availabilities and concentrations and physical properties is crucial for minimizing the climatic impacts of rice agriculture. We examined soil nutrients and other properties, greenhouse-gas production and their relationships in 26 paddy fields throughout the province of Fujian in China, one of the most important provinces for rice production. High P and K concentrations, contents and availabilities were correlated with low rates of $CO_2$ production, whereas high C and N contents were correlated with high rates of $CH_4$ production. Mean annual precipitation (MAP) and rates of gas production were not clearly correlated, at least partly due to the management of flooding that can mask the effect of precipitation. Higher mean annual temperatures and soil Fe contents favored the production of $N_2O$. C, N, P and K concentrations and their ratios, especially the C:K and N:K ratios, and P availability were correlated with $CO_2$ and $CH_4$ production across the province, with higher C:K and N:K ratios correlated positively with increased $CO_2$ production and available P correlated negatively with $CH_4$ production. A management strategy to avoid excessive C accumulation in the soil and to increase P availability and decrease available Fe contents would likely decrease the production of greenhouse gases.

Keywords: Paddy field; $CH_4$ flux; $N_2O$ flux; Greenhouse gases; Elemental stoichiometry; Diurnal variation; Nitrogen; Soil nutrients; Phosphorus; Seasonal variation; Warming

## 1. Introduction

Rice currently feeds more than 50% of the global population (Haque et al., 2015), but production will need to increase by 40% by the end of 2030 to meet the demand for food from the growing population worldwide (FAO, 2009). Sustaining soil fertility and increasing the rice yields are therefore of utmost importance. Increased nutrient supply can stimulate the growth and grain yield of rice plants (Ali et al., 2008; Wang et al., 2014a). Paddy fields, though, are very important sources of greenhouse gases (GHGs), especially methane ($CH_4$) and nitrous oxide ($N_2O$) (Myhre et al., 2013), so minimizing the release of these very potent GHGs from paddies, thus mitigating their adverse impacts on climate change, are also of utmost importance. $CO_2$, $CH_4$ and $N_2O$ are the most important GHGs, contributing about 80% of the current global radiative forcing (Myhre et al., 2013).

Improving the availability of soil nutrients in paddy fields while decreasing GHG production, or at least not increasing it, is an outstanding challenge. Reaching this objective requires understanding the relationships between soil nutrients and GHG production and emission. Most current studies relating greenhouse gas emissions from paddy fields with soil properties have been conducted in a single location by applying different treatments that modify soil properties. Several strategies of rice crops management, from fertilizer and herbicide application to distinct straw or water management, have been proved to increase rice yield without increasing or decreasing CO2 (Li et al., 2005, 2013; Liu et al., 2016), CH4 (Jiang et al., 2015; Launio et al., 2016; Takakai et al., 2017; Trinh et al., 2017; Wang et al., 2016; Zhang et al., 2016) and/or N2O (Jiang et al., 2015; Launio et al., 2016; Trinh et al., 2017; Wang et al., 2016; Zhang et al., 2016) emissions. Nitrogen applications have been mostly positively related to N2O emissions in wetland areas and paddy soils (Ma et al., 2007; Liu and Grover, 2009;

Wang et al., 2013; Parn et al., 2015). The addition of great fertilization dose of N and/or P fertilizers increases $CO_2$, $CH_4$ and $N_2O$ soil emission from paddy soils (Ma et al., 2007; Mon et al., 2014; Riya et al., 2015; Hu et al., 2015). However, some studies have no detected relationships between N fertilization and $CO_2$, $CH_4$ and $N_2O$ soil emissions (Xie et al., 2010; Morris et al., 2017) or even decreases of soil $CH_4$ emissions (Cai et al., 2007; Yao et al., 2012; Li et al., 2012), or different effects increasing or decreasing $CH_4$ and $N_2O$ emissions depending on the type of N-fertilizer used (Trinh et al., 2017). Most studies, however, have found a significant relationships among fertilization and crop management strategies with soil N, P and K contents and availability and thereafter with soil greenhouse gas emissions (Adhya et al., 1998; Iqbal et al., 2009; Dong et al., 2011; Zhu et al., 2011; Zheng et al., 2013; Zhao et al., 2015; Bordoloi et al., 2016; Dossou-Yovo et al., 2016; Jiang et al., 2016; Sheng et al., 2016; Wang et al., 2017). Thus there is a general consensus that different rice crop management change the rates of greenhouse gas emissions, and that this change is related to the changes in soil N, P and K concentrations produced by the different management strategies. However, the research of possible relationships among C, N and K contents and stoichiometries with the shifts of the soil $CO_2$, CH4 and $N_2O$ emissions have not been studied in paddy fields. We investigated these possible general links by analyzing the $CO_2$, $CH_4$ and $N_2O$ emissions from 26 paddy soils spread through Fujian province (China) under different long-term fertilization managements and under distinct environments from coastal wetland areas to mountain terraces. In many of those studies the links between different general soil traits such as pH (Wang et al., 2017), Eh, (Fan et al., 2016; Wang et al., 2015 and 2017) or salinity (Olsson et al., 2015) with greenhouse gas emissions have been observed. We also included the shifts in these general soil variables and their potential links with crop management, soil nutrient status and greenhouse gas emissions in our study. We hypothesize that different soil C, nutrient content, salinity or pH taken one-by-one or better in combination should explain a significant part of the differences of gas emissions among sites. This knowledge should be used for improving the management strategies of soils conditions towards more favorable soil traits to hinder as much as possible GHG emissions.

China has the second largest area of rice cultivation in the world, and the associated GHG emissions account for about 40% of the total agricultural sources of GHGs. Sixty percent of the Chinese population depends on rice-based food, so the protection of China's rice production for food security is important (Zhu, 2006). Ninety percent of the paddies in China are in the subtropics, such as in Fujian, Jiangxi and Hunan Provinces. Developing effective strategies to increase the cost-effectiveness of rice agriculture, enhancing soil nutrients and mitigating GHG emissions from paddies in subtropical China to minimize future problems of food shortage and adverse climate change are thus of national and global importance. The potential production of $CO_2$, $CH_4$ and $N_2O$ for all of Fujian Province, calculated as the average production per unit area (production rate x soil depth x days of the year (365 d in 2014)) × area of the paddy fields), was 2747.59, 9.85 and 3.10 Mg $ha^{-1}$ $y^{-1}$, respectively. An understanding of the relationships between soil properties and gas emissions can thus help us to modify the current strategies of crop management to decrease gas emissions.

We pursued this objective by: (1) determining the rates of $CO_2$, $CH_4$ and $N_2O$ production in 26 paddy fields in Fujian province, (2) measuring the levels of soil nutrients and other soil traits in the paddy fields and (3) assessing the relationships between GHG production and the various soil traits.

## 2. Material and methods

*2.1. Study area*

This study was conducted in Fujian province in southeastern China, between the mid-subtropical and the southern subtropical zones (Fig. 1). Fujian province has an area of $1.21 \times 10^5$ $km^{-2}$ (Hong and Wang, 2009). The climate is relatively warm and wet with a mean annual temperature of 17-21 °C and a mean annual precipitation of 1000-2200 mm (Hong and Wang, 2009). Many paddy fields are distributed in coastal and inland areas, and we selected 26 of these paddy fields (Fig. 1).

*2.2. Collection and measurement of soil samples*

Soil samples were collected in August 2014 from three replicate plots in 26 randomly selected paddy fields. Soil samples were collected at the same time with a small core sampler (length and diameter of 0.5 and 0.1 m, respectively) from the 0-15 cm layer, representing the plowed layer. A total of 78 samples (26 fields × 1 soil layer × 3 replicates) were thus collected. The samples used for all analyses were mixed core samples. The samples were air-dried, and roots and visible plant remains were removed.

Total C and N concentrations were measured using a Vario EL III Elemental Analyzer (Elementar Scientific Instruments, Hanau, Germany, Wang et al., 2015a; Wang et al., 2016a). Amounts of labile organic carbon (LOC) were determined by digestion with 333 mM $KMnO_4$ (Wang et al., 2015b). Available N concentration was determined by the diffusion method using alkaline hydrolysis (Lu, 1999). Total P concentrations were determined by Mo-Sb colorimetry (Lu, 1999; Ruban et al., 1999). Available P concentration was determined by extraction with 0.5 mol $L^{-1}$ $NaHCO_3$ and then measured by Mo-Sb colorimetry (Lu, 1999; Ruban et al., 1999) using the UV-2450 spectrophotometer (Shimadzu Scientific Instruments, Kyoto, Japan) at a wavelength of 700 nm (Wang et al., 2016b). Total K concentration was determined by FP 640 flame photometry (Shanghai Electronic Technology Instruments, Shanghai, China, Wang et al., 2014b). Available K concentration was determined by extraction with 1 mol $L^{-1}$ $NH_4Ac$ and then measured by FP 640 flame photometry (Shanghai Electronic Technology Instruments, Shanghai, China, Wang et al., 2014b). Total Fe concentration was determined by digesting fresh soil with 1M HCl, extracting the ferrous ions using 1,10-phenanthroline and measuring the concentration by spectrometrically (Lu, 1999; Wang et al., 2014a). Ferric Fe concentration was calculated by subtracting the ferrous Fe concentration from the total Fe concentration (Wang et al., 2014a).

Soil $CO_2$, $CH_4$ and $N_2O$ production was determined by anaerobic incubation (Wang et al., 2010; Wang et al., 2015a). Thirty grams of fresh soil of the three mixed core soil samples of each site were placed into 120-mL incubation bottles, and two volumes of distilled water were added. The bottles were purged with $N_2$ for 2 min to replace the $O_2$ and were then sealed with a rubber stopper and incubated at 20 °C for 3 d. Five milliliters of gas were extracted from the headspaces four times each day.

$CO_2$, $CH_4$ concentrations in the headspace air samples were determined by gas chromatography (Shimadzu GC-2010, Kyoto, Japan), conducted with a Porapak Q column (2 m length, 4 mm OD, 80/100 mesh, stainless steel column). $N_2O$ concentrations in the headspace air samples was determined by gas chromatography (Shimadzu GC-2014, Kyoto, Japan), conducted with a Porapak Q column (2 m length, 4 mm OD, 80/100 mesh, stainless steel column). A methane conversion furnace, flame ionization detector (FID) and electron capture detector (ECD) were used for the determination of the $CO_2$, $CH_4$ and $N_2O$ concentrations, respectively. The operating temperatures of the column, injector and detector were adjusted to 45, 100 and 280 °C, respectively, for the determination of the $CO_2$ concentration, to 70, 200 and 200 °C, respectively, for the determination of the $CH_4$ concentration and to 70, 200 and 320 °C, respectively, for the determination of the $N_2O$ concentration. Helium (99.999% purity) was used as a carrier gas (30 mL $min^{-1}$), and a make-up gas (95% argon and 5% $CH_4$) was used for the ECD. The gas chromatograph was calibrated before and after each set of measurements using 503, 1030 and 2980 μL $CO_2$ $L^{-1}$ in He; 1.01, 7.99 and 50.5 μL $CH_4$ $L^{-1}$ in He and 0.2, 0.6 and 1.0 μL $N_2O$ $L^{-1}$ in He (CRM/RM information center of China) as primary standards (Wang et al., 2015c, d). We use linear equation and calculation of $CO_2$, $CH_4$ and $N_2O$ production.

Other soil variables were also analyzed. Bulk density was measured from three 5 × 3 cm cores (Wang et al., 2016b), pH was measured with a pH meter (IQ Scientific Instruments, Carlsbad, USA, Wang et al., 2016b) 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 Analyser (Malvern Scientific Instruments, Malvern, UK, Wang et al., 2016b).

*2.3. Determination of soil C and nutrient contents*

The C, N, P, K and Fe contents in the 0-15 cm soil profiles were estimated by (Mishra et al., 2010):

$$C_S = \sum_{j=1}^{n} c_{\mathrm{m}} \times \rho_{\mathrm{b}} \times D$$

where $C_S$ is the C, N, P, K or Fe content (kg $m^{-2}$), $j$ is the soil-depth interval (1, 2, … n), $C_m$ is the C, N, P, K or Fe concentration (g $kg^{-1}$), $\rho_b$ is the soil bulk density (kg $m^{-3}$), $D$ is the thickness of each soil layer (m) and $n$ is the number of soil layers.

*Determination of soil $CO_2$, $CH_4$ and $N_2O$ production*

We used linear regression calculation for $CO_2$, $CH_4$ and $N_2O$ production as (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 $CO_2$, $CH_4$ or $N_2O$ production ($\mu g^{-1}$ $g^{-1}$ $d^{-1}$), $dc/dt$ is the recorded change in the mixing ratio of C ($CO_2$, $CH_4$ and $N_2O$) in the headspace over time (mmol $mol^{-1}$ $d^{-1}$), $V_H$ is the volume of the headspace (L), $Ws$ 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).

*2.4. Statistical analyses*

The relationships of each soil variable with $CO_2$, $CH_4$ and $N_2O$ production were determined by simple regressions using Statistica 6.0 (StatSoft, Inc. Tulsa, USA).

We analyzed the effects of multiple soil variables as fixed factors on the production rates of the three GHGs using general linear models with and without spatial correlation. We also analyzed the effect of location on all variables using a linear mixed model, with location (coastal or inland) as a fixed factor and site as a random factor. We used the "*nlme*" (Pinheiro et al., 2016) and "*lme4*" (Bates et al., 2015) R packages with the "*lme*" the "*lmer*" functions, respectively. We chose the best model for each dependent variable by following Akaike's information criterion (AIC). We used the MuMIn R (Barton, 2012) package in the mixed models to estimate the percentage of variance explained by the model.

We identified the soil variables that were correlated best with the rates of gas production by dividing the study sites into three groups for each emitted gas: those with low, medium and high rates of gas production. We then performed multivariate statistical analyses using a discriminant functional analysis (DFA) to determine the importance of the various combinations of variables in the grouping of the sites by the three groups of gas-production rates. The final model chosen was that where all variables used for grouping were significant. The DFAs were performed using Statistica 6.0 (StatSoft, Inc. Tulsa, USA). The C:N, C:P, C:K, N:P, N:K and P:K ratios and the LOC:available-N, LOC:available-P, LOC:available-K, available-N:available-P, available-N:available-K and available-P:available-K ratios were calculated as mass ratios.

We analyzed the relationships of overall soil elemental composition and stoichiometry with climate (mean annual precipitation and temperature, MAP and MAT) and gas production using a principal component analysis (PCA) of all elemental and stoichiometric variables. The scores of the two first PC axes were then correlated by linear regressions with MAP, MAT and the production rates of the GHGs. The PCAs and regression analyses were performed using Statistica 6.0 (StatSoft, Inc. Tulsa, USA).

## 3. Results

### *3.1. GHG production and soil traits*

Soil $CO_2$ production was correlated positively with soil total C and N concentrations and MAP and negatively with total K concentration, available K concentration, available P concentration and pH (Table 1). $CH_4$ production was not correlated with MAP or pH, but $CO_2$ production was correlated positively with total C and N concentrations and negatively with total K concentration and available K concentration (Table 1). $CH_4$ production was negatively correlated with $Fe^{3+}$ concentration (Table 1). $N_2O$ production, however, was poorly correlated with the soil variables, only negatively with the LOC:available-N ratio and positively with silt concentration and MAT (Table

1).

The best linear models of the combinations of variables influencing the rates of gas production showed that total and available P concentrations, total K concentrations and pH explained 44% of the total variance of the rates of $CO_2$ production (Table 2). Total C and N concentrations and the total-N:K ratio explained 48% of the total variance of the rates of $CH_4$ production (Table 2). $Fe^{3+}$ concentration, MAT and the available-C:N ratio explained 24% of the total variance of the rates of $N_2O$ production (Table 2).

The DFA with total K concentration, available P and K concentrations, the total-P:K ratio and pH as independent continuous variables significantly separated all three groups of $CO_2$ production rates (Tables S1 and S2, Fig. 2). The DFA with total C and N concentrations, LOC concentration, and the total-N:K and C:K ratios as independent continuous variables significantly separated all three groups of $CH_4$ production rates, although the separation between the groups with low and medium rates was marginally significant ($P$=0.054) (Tables S3 and S4, Fig. 3). The DFA with LOC, the available-C:N ratio and MAT as independent continuous variables significantly separated all three groups of $N_2O$ production rates (Tables S5 and S6, Fig. 4).

### *3.2. Differences in soil variables and rates of GHG production between coastal and inland paddy fields*

On average, available P content, pH and salinity were higher and the available-C:P ratio and rate of $CO_2$ production were lower in the coastal than the inland paddy fields (Table 3). Moreover $CH_4$ emissions were marginally lower in coastal than in inland rice crops and available-$K^+$ content was higher in coastal than inland rice crops (Table 3). The coastal paddy fields also had, on average, a lower MAP than the inland paddy fields.

## 4. Discussion

### *4.1. Soil C and nutrient contents and stoichiometries and GHG emissions*

High soil P and K concentrations, contents and availabilities were correlated with lower rates of $CO_2$ production, consistent with reports that soil P and K were not limiting nutrients for $CO_2$ production rates. Previous studies have shown that N but not P or K was the most common limiting nutrient in Fujian paddy fields. Olsson et al. (2005) also reported that the fertilization (including N, P, K, Mg and micronutrients) of boreal forests decreased both autotrophic and heterotrophic soil respiration. But contrarily higher amounts of soil available-P have been positively related with soil microbioal heterotrophic and autotrophic respiration and thus with larger soil CO2 efflux (Wright and Reddy, 2001). Our study is the first to report a negative relationship between P and K and $CO_2$ production in paddy fields on a large geographical scale. Lukito et al (1998) observed that exist a critical soil microbial P concentration that it is the optimum to achieve the maximum microbial biomass synthesis under certain soil conditions, above P-concentrations tend to decrease microbial activity. P is not limiting in soil and its availability is quiet high in the studied area (Wang et al., 2015b). Soil-K has been correlated with increases in salinity (Borgognone et al., 2014), and high salinity can decrease microbial biomass (Elmajdoub and Marschner, 2013) and thus soil respiration and $CO_2$ production (Mavi et al., 2012; Ondrasek et al., 2012). Lower available-P:K ratios in our study were correlated with high $CO_2$ production, also highlighting the relationships between the ratios of available nutrients and gas production.

K and P are important elements for the growth of methanogenic microbes, but our study found that lower amounts of P and K could be correlated with high $CH_4$ production. The use efficiencies of P and K in the paddy fields of our study area (Fujian province) are low (Zhu, 2006), and these nutrients, especially P, are retained in the soil, and a high P availability can inhibit methane production via acetate fermentation (Conrad and Klose, 2000). In most of the study area, methane production via acetate fermentation is significantly higher than methane production via $CO_2$ reduction (Avery et al., 2003). Low amounts of P can thus be associated with high $CH_4$ production. Increases in K will increase salinity (Borgognone et al., 2014), and high salinity will decrease microbial activity (Elmajdoub and Marschner, 2013) and thus methane production (Mavi et al., 2012; Ondrasek et al., 2012). Lower K concentrations can therefore lead to high $CH_4$ production.

High soil C and N contents were correlated with high rates of $CH_4$ production in our study. The paddy fields in Fujian province are frequently limited by C and N (Wang et al., 2015b). Soil organic carbon and soil N concentrations were positively correlated with $CO_2$ and $CH_4$ soil efflux. These results are in agreement with most previous reports. Low N availability in soils can constrain soil microbial activity and thus soil $CO_2$ efflux (Inselbacher et al., 2011; Gavrilenko et al., 2011; Christiansen et al., 2012). Soil C and N concentrations have been positively related with Methanobacteria abundance in soils (Zhu et al., 2010). In a previous study in the same study area, $CH_4$ emission increased in the rice growing season after the addition of rice straw, coinciding with the maximum temperature, and $CH_4$ production was limited by the amount of C substrates (Wang et al., 2015c). An increase in soil N concentration will increase $CH_4$ production and emissions, which could resulted even more probably given to the N limitation that has also been observed in our studied area (Wang et al., 2015b). An increase in N can also have a positive effect on $CH_4$ emission by promoting the input of photosynthetically derived C into soil organic carbon pools, because N can promote microbial activity and population sizes, increasing the rate of root decomposition and the release of C into the soil. N will also increase plant growth, which will add C to the SOC pools by increasing root biomass (Ge et al., 2015). Newly photosynthesized C can contribute up to 52% of the $CH_4$ production and emission from paddy soils by the exudation of LOC from the roots to the rhizosphere for methanogenesis, hence promoting $CH_4$ production and emission from the soils (Minoda et al., 1996; Lai et al., 2014). The other 48% is emitted from old soil C (Minoda et al., 1996).

We have observed that soils with more Fe content had higher potential to $N_2O$ emission in lab conditions. The rate of $N_2O$ production is correlated with Fe content (Maag and Vinther, 1996), because Fe is an important factor controlling $N_2O$ production (Zhu et al., 2013). High $Fe^{3+}$ concentrations could enhance the release of $N_2O$ to the atmosphere by inhibiting the enzymatic reduction of $N_2O$ in soils (Huang et al., 2009). There was a negative relationships between LOC and the potential rates of $N_2O$ emission and also with low available C:N ratio. Thus, the overall results suggest that soils with lower soil LOC, and lower C:N, when moreover, contain large amounts of iron are the soils with the largest potential to emit $N_2O$. Negative relationship between soil C:N ratio and the rates of $N_2O$ emission have been previously observed (Toma and Hatano, 2007; Stevenson et al., 2010).

Our analyses strongly suggest that the availabilities and imbalances of soil nutrients, especially lower total-C:K and N:K ratios and a higher P availability, could decrease $CO_2$ and $CH_4$ production across Fujian province. A management strategy for attaining better nutrient ratios and P availability should thus be investigated. Moreover, denitrification may be the most important pathway of $N_2O$ production in paddy fields, so N fertilizer in the form of $NH_4^+$ should be added during the rice growing season.

The total C, total N, total P, total K, total Fe, LOC, available N, available P, available K, $Fe^{2+}$ and $Fe^{3+}$ contents for all of Fujian Province calculated as the average content × area of the paddy field were 370 836.90, 37 887.91, 4654.65, 110 800.49, 114 792.02, 126 261.33, 796.76, 311.65, 5461.03, 37 534.26 and 77 257.76 Mg $ha^{-1}$, respectively. The ratios (on a mass basis) between LOC and N, P and K were 159, 406 and 23, respectively, indicating clear stoichiometric imbalances, especially considering the very high LOC:N and LOC:P ratios for microbes and plants. These results imply an excess of labile C in the soil, with positive correlations between LOC and $CO_2$ production ($P<0.05$), between the available-C:N ratio and $CH_4$ production ($P<0.05$) and between the available-C:N ratio and $CO_2$ production ($P<0.1$).”

*4.2. Climate and gas emissions*

Site MAP had negative relationships with soil available-P concentration and C:P and N:P ratios (data not shown). In turn, available P concentration and available P content were negatively correlated with $CO_2$ production. Site MAP can increase P leaching and soil $CO_2$ emissions in the periods with no flooding. Larger total $CO_2$ emissions during the rice crop cycle are related with higher natural precipitation, lower soil pH and P-availability but higher P use efficiency (higher C:P ratios). However, the patterns between climate and rate of gas production were not clear in part due to the management of flooding that masks the changes due to climatic conditions. We have observed a positive relationship between site MAT and $N_2O$ production rates. An increase in the rate of $N_2O$ production due to climatic warming has been reported in a previous study and is associated with nitrification and denitrification, which are the main pathways of $N_2O$ production (Dobbie and Smith, 2001). Site MAT was negatively related with LOC that in turn is related with lower potential rates of $N_2O$ emission. Thus, overall, the results suggest that soils with high MAT are related with lower soil LOC, and that when there are large iron contents, also with a largest potential to emit $N_2O$.

*4.3. Comparison of coastal and inland fields*

On average, pH and salinity were higher in the coastal than the inland paddy fields, because the fields were flooded with water from coastal wetlands (Wang et al., 2014b) and the inland paddy fields were flooded with water from riparian wetlands (Wang et al., 2015a). The available-P content was also higher in the coastal than the inland paddy fields, due to the higher salinity and pH (Wang et al., 2016b). High salinity and pH will increase P availability by the displacement of phosphate from ligand exchange sites by Ca, K, Mg and Na cations (Roy et al., 1971; Lee et al., 2004) and/or by decreasing phosphate sorption on soil colloids (Shariatmadari et al., 1999). The coastal paddy fields in our study also had less LOC, so the available-C:P ratio was lower. The rates of $CO_2$ production were lower in the coastal than the inland paddy fields, consistent with the larger LOC concentration in the coastal paddy fields. LOC substrates are very important for the production of $CO_2$, and as the amount of C increases, the amount and activity of soil microbes will also increase, thereby promoting $CO_2$ production (Curtin et al., 1998; Cleveland et al., 2007). $CO_2$ production in our study was thus higher in the inland paddy fields, and also not significant $CH_4$ emissions were also lower in coastal weatlands also consistently with DFA analysis that relates $CH_4$ emissions positively with LOC, and soil C and N concentrations (Figure 3). The rates of $CO_2$ production were lower in the coastal than the inland paddy fields, also due to the higher salinity in the coastal paddy fields. $CO_2$ production has previously been negatively correlated with soil salinity (Setia et al., 2010; Setia et al., 2011); high salinity inhibits the growth and activity of soil microorganisms due to osmotic stress and inhibits $CO_2$ production (Rietz and Haynes, 2003). These relationships of the emissions of $CO_2$ and also $CH_4$ related to soil C, N, P and K contents and availability are consistent with the linear and DFA models. A consistent relationship between higher soil P and K availability and low LOC and soil C and N contents with low $CO_2$ and $CH_4$ emissions arises from all these analyses. All these results are consistent with the fact that inland rice crops had larger time for growth and more P and K is accumulate in plant, thus P and K could be lees in soil. Moreover, in inland rice crops, more rice straw was returned to crop, and Azolla sp. and Astragalus sinicus are planted in no rice growth period. Thus, with more straw input and with the growth of a leguminous $N_2$-fixing (Astragalus) each year, we must expect higher soil C and N concentrations. Moreover, during no rice period there is no management in coastal rice crops. In the case of $N_2O$ emissions, the differences are not significant between coastal and inland rice crop sites, despite on average the $N_2O$ emissions were higher in coastal than in Inland areas This was consistent with lower soil LOC and higher available-C:N and MAT (related to higher $N_2O$ emission rates, such as above discussed) observed in coastal than in inland rice crops. Coastal rice croplands also tend to have lower MAPs than inland croplands. The inland paddy fields in our study were mainly distributed in the western areas of Fujian province, where the altitude is higher, especially in the Wuyi Mountains, and the coastal paddy fields were mostly in eastern areas with low hills and plains. Mean annual precipitation thus also varied, ranging from 1200 mm on the coast to 2000 mm inland.

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## Conflicts of Interest

The authors declare no conflicts of interest.

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## Tables

**Table 1**

Regression statistics for the rates of $CO_2$, $CH_4$ and $N_2O$ production associated with the various soil variables. We controlled the false discovery rate of multiple correlations (Benjamini and Hochberg, 1995). Significant statistics ($P$<0.004) after applying the Benjamini and Hochberg correction are highlighted in bold type.

| Soil variable | Greenhouse-gas production | | |
|---|---|---|---|
| | Rate of $CO_2$ production | Rate of $CH_4$ production | Rate of $N_2O$ production |
| Total [C] | ***R*=0.34 *P*=0.002** | ***R*=0.52 *P*<0.0001** | *R*=0.023 *P*=0.84 |
| Total [N] | ***R*=0.39 *P*<0.0001** | ***R*=0.57 *P*<0.0001** | *R*=0.012 *P*=0.91 |
| Total [P] | *R*=0.21 *P*=0.071 | *R*=0.19 *P*=0.089 | *R*=0.15 *P*=0.18 |
| Total [K] | ***R*=-0.35 *P*=0.002** | ***R*=-0.33 *P*=0.003** | *R*=0.10 *P*=0.36 |
| Total [Fe] | *R*=0.16 *P*=0.168 | ***R*=-0.34 *P*=0.002** | *R*=0.14 *P*=0.23 |
| [LOC] | ***R*=0.29 *P*=0.010** | ***R*=0.33 *P*=0.004** | ***R*=-0.26 *P*=0.020** |
| Available [N] | *R*=0.13 *P*=0.27 | *R*=-0.17 *P*=0.14 | *R*=0.13 *P*=0.25 |
| Available [P] | ***R*=-0.33 *P*=0.003** | *R*=-0.0094 *P*=0.94 | *R*=0.22 *P*=0.050 |
| Available [K] | *R*=-0.057 *P*=0.620 | *R*=-0.071 *P*=0.54 | *R*=0.065 *P*=0.57 |
| $[Fe^{2+}]$ | *R*=0.070 *P*=0.55 | *R*=-0.11 *P*=0.34 | *R*=0.23 *P*=0.044 |
| $[Fe^{3+}]$ | *R*=0.17 *P*=0.14 | ***R*=-0.38 *P*<0.0001** | *R*=0.084 *P*=0.47 |
| C:N ratio | *R*=0.12 *P*=0.29 | *R*=0.14 *P*=0.22 | *R*=0.084 *P*=0.46 |
| C:P ratio | *R*=0.12 *P*=0.28 | *R*=0.18 *P*=0.13 | *R*=-0.062 *P*=0.59 |

| | | | |
|---|---|---|---|
| C:K ratio | **_R_=0.34 _P_=0.002** | **_R_=0.67 _P_<0.0001** | _R_=-0.081 _P_=0.48 |
| N:P ratio | _R_=0.090 _P_=0.44 | _R_=0.14 _P_=0.23 | _R_=-0.11 _P_=0.35 |
| N:K ratio | **_R_=0.35 _P_=0.002** | **_R_=0.67 _P_<0.0001** | _R_=-0.091 _P_=0.43 |
| P:K ratio | **_R_=0.33 _P_=0.003** | **_R_=0.59 _P_<0.0001** | _R_=-0.053 _P_=0.64 |
| LOC:N ratio | _R_=0.20 _P_=0.073 | **_R_=0.30 _P_=0.009** | **_R_=-0.31 _P_=0.006** |
| LOC:P ratio | _R_=0.17 _P_=0.14 | _R_=0.065 _P_=0.57 | _R_=-0.14 _P_=0.23 |
| LOC:K ratio | _R_=0.065 _P_=0.60 | _R_=0.087 _P_=0.45 | _R_=-0.13 _P_=0.25 |
| Available N:P ratio | _R_=0.13 _P_=0.26 | _R_=-0.059 _P_=0.61 | _R_=-0.040 _P_=0.72 |
| Available N:K ratio | _R_=0.13 _P_=0.26 | _R_=-0.059 _P_=0.61 | _R_=-0.040 _P_=0.73 |
| Available P:K ratio | **_R_=-0.32 _P_=0.004** | _R_=-0.0020 _P_=0.99 | _R_=0.16 _P_=0.17 |
| Bulk density | _R_=-0.18 _P_=0.11 | _R_=-0.14 _P_=0.21 | _R_=-0.072 _P_=0.530 |
| pH | **_R_=-0.30 _P_=0.007** | _R_=-0.15 _P_=0.20 | _R_=0.11 _P_=0.34 |
| Salinity | _R_=-0.10 _P_=0.37 | _R_=-0.069 _P_=0.55 | _R_=-0.0058 _P_=0.96 |
| Clay content | _R_=-0.11 _P_=0.33 | _R_=0.059 _P_=0.61 | _R_=0.097 _P_=0.40 |
| Silt content | _R_=0.20 _P_=0.074 | _R_=0.066 _P_=0.57 | **_R_=0.29 _P_=0.011** |
| Sand content | _R_=-0.13 _P_=0.245 | _R_=-0.070 _P_=0.54 | _R_=-0.26 _P_=0.022 |
| MAT(2009-2013) | _R_=-0.19 _P_=0.093 | _R_=-0.086 _P_=0.46 | **_R_=0.31 _P_=0.006** |
| MAP (2009-2013) | **_R_=0.30 _P_=0.008** | _R_=0.13 _P_=0.27 | _R_=-0.18 _P_=0.11 |

**Table 2**

The best general linear models (lowest AICs), with soil variables as independent continuous variables and the rates of gas production as dependent variables. Models were tested with and without spatial autocorrelation.

<table>
<tr><td>Variable</td><td colspan="5"></td><td>Total model statistics</td></tr>
<tr><td rowspan="2">$CO_2$ production rate</td><td>Soil total [C]</td><td>Soil total [N]</td><td>Soil total [K]</td><td>Soil P-availability</td><td>MAP</td><td rowspan="2">$R^2$=0.38<br>$F$=9.4<br>$P$<0.0001</td></tr>
<tr><td>$F$= 13.4<br>$P$<0.001</td><td>$F$=4.11<br>$P$=0.046</td><td>$F$=6.35<br>$P$=0.0141</td><td>$F$=15.3<br>$P$<0.001</td><td>$F$=4.25<br>$P$=0.043</td></tr>
<tr><td rowspan="2">$CH_4$ production rate</td><td colspan="2">Soil total [C]</td><td colspan="2">Soil total [N]</td><td>Soil [Fe]</td><td rowspan="2">$R^2$=0.40<br>$F$=15.5<br>$P$<0.0001</td></tr>
<tr><td colspan="2">$F$=32.5<br>$P$<0.0001</td><td colspan="2">$F$=6.85<br>$P$<0.01</td><td>$F$=6.60<br>$P$<0.01</td></tr>
<tr><td rowspan="2">$N_2O$ production rate</td><td colspan="2">Soil LOC</td><td colspan="2">Silt (%)</td><td>Soil [Fe]</td><td rowspan="2">$R^2$=0.25<br>$F$=7.0<br>$P$<0.001</td></tr>
<tr><td colspan="2">$F$=6.39<br>$P$=0.014</td><td colspan="2">$F$=7.51<br>$P$<0.01</td><td>$F$=6.76<br>$P$=0.011</td></tr>
</table>

**Table 3**

Linear mixed models of the relationships of paddy-field location with soil variables and rates of gas production. Plot site was determined by location (coastal versus inland) as a fixed factor and each site as a random factor. The dependent variables explaining a significant proportion ($P$<0.05) of the total variance are marked with an asterisk (*). $R^2$ values in bold type highlight the model with the lowest AIC when comparing models with and without spatial correlation for each dependent variable. $R^2$m is $R^2$ value of fixed factors. $R^2$c = value of fixed factors. $R^2$m = value of fixed plus random factors.

| | Mixed linear model: Y = location (fixed) + site (random) | | | | Coastal areas | Inland areas |
|---|---|---|---|---|---|---|
| | Location (inland versus coastal) | Model $R^2$ Without autocorrelation | Model $R^2$ With autocorrelation | Model comparison (with versus without spatial autocorrelation) | Mean ± S.E. | Mean ± S.E. |
| Total C concentration (g $kg^{-1}$) | F=0.87 $P$=0.36 | **$R^2$m=0.031 $R^2$c=0.94** | $R^2$m=0.031 $R^2$c=0.94 | $P$=0.99 | 17.3 ± 1.6 | 20.2 ± 0.9 |
| Total N concentration (g $kg^{-1}$) | F=0.33 $P$=0.35 | **$R^2$m=0.032 $R^2$c=0.92** | $R^2$m=0.032 $R^2$c=0.92 | $P$=0.99 | 1.80 ± 0.11 | 2.05 ± 0.08 |
| Total P concentration (g $kg^{-1}$) | F=0.75 $P$=0.39 | $R^2$m=0.025 $R^2$c=0.81 | **$R^2$m=0.027 $R^2$c=0.74** | $P$=0.015 | 0.268 ± 0.023 | 0.230 ± 0.014 |
| Total K concentration (g $kg^{-1}$) | F=0.73 $P$=0.40 | $R^2$m=0.028 $R^2$c=0.99 | **$R^2$m=0.027 $R^2$c=0.98** | $P$=0.0002 | 6.51 ± 0.72 | 5.43 ± 0.40 |
| Total Fe concentration (g $kg^{-1}$) | F=1.09 $P$=0.31 | **$R^2$m=0.039 $R^2$c=0.94** | $R^2$m=0.039 $R^2$c=0.94 | $P$=0.99 | 7.03 ± 0.74 | 5.54 ± 0.42 |
| LOC concentration (mg $kg^{-1}$) | F=1.45 $P$=0.24 | **$R^2$m=0.045 $R^2$c=0.74** | $R^2$m=0.044 $R^2$c=0.74 | $P$=0.87 | 5345 ± 452 | 6972 ± 484 |
| Available N concentration (mg $kg^{-1}$) | F=0.051 $P$=0.82 | **$R^2$m=0.0019 $R^2$c=0.90** | $R^2$m=0.0019 $R^2$c=0.90 | $P$=0.89 | 44.6 ± 6.0 | 40.9 ± 5.3 |
| Available P concentration (mg $kg^{-1}$) | F=3.66 $P$=0.068 | **$R^2$m=0.12 $R^2$c=0.98** | $R^2$m=0.12 $R^2$c=0.97 | $P$=0.15 | 26.7 ± 5.3 | 11.5 ± 1.9 |
| Available K concentration (mg $kg^{-1}$) | F=3.67 $P$=0.079 | $R^2$m=0.073 $R^2$c=0.44 | **$R^2$m=0.077 $R^2$c=0.41** | $P$=0.10 | 360 ± 42 | 259 ± 18 |
| $Fe^{2+}$ concentration (g $kg^{-1}$) | F=0.39 $P$=0.54 | **$R^2$m=0.014 $R^2$c=0.93** | $R^2$m=0.014 $R^2$c=0.93 | $P$=0.99 | 2.17 ± 0.26 | 1.89 ± 0.12 |
| $Fe^{3+}$ concentration (g $kg^{-1}$) | F=1.11 $P$=0.30 | **$R^2$m=0.04 $R^2$c=0.94** | $R^2$m=0.04 $R^2$c=0.94 | $P$=0.99 | 4.86 ± 0.55 | 3.65 ± 0.35 |
| C:N ratio | F=0.32 $P$=0.58 | **$R^2$m=0.012 $R^2$c=0.99** | $R^2$m=0.012 $R^2$c=0.99 | $P$=0.62 | 9.34 ± 0.40 | 9.79 ± 0.23 |
| C:P ratio | F=1.75 $P$=0.20 | $R^2$m=0.061 $R^2$c=0.93 | **$R^2$m=0.062 $R^2$c=0.92** | $P$=0.035 | 74.6 ± 8.6 | 106 ± 8 |
| C:K ratio | F=0.86 $P$=0.36 | **$R^2$m=0.032 $R^2$c=0.99** | $R^2$m=0.032 $R^2$c=0.99 | $P$=0.99 | 3.33 ± 0.46 | 5.27 ± 0.69 |
| N:P ratio | F=1.48 $P$=0.24 | $R^2$m=0.051 $R^2$c=0.90 | **$R^2$m=0.052 $R^2$c=0.87** | $P$=0.0081 | 7.95 ± 0.90 | 10.6 ± 0.7 |
| N:K ratio | F=0.75 $P$=0.39 | **$R^2$m=0.028 $R^2$c=0.99** | $R^2$m=0.028 $R^2$c=0.99 | $P$=0.99 | 0.347 ± 0.040 | 0.538 ± 0.074 |
| P:K ratio | F=0.12 $P$=0.73 | $R^2$m=0.005 $R^2$c=0.97 | $R^2$m=0.0046 $R^2$c=0.97 | $P$=0.66 | 0.051 ± 0.005 | 0.059 ± 0.008 |
| Available-C:N ratio | F=1.40 $P$=0.25 | **$R^2$m=0.035 $R^2$c=0.51** | $R^2$m=0.035 $R^2$c=0.51 | $P$=0.99 | 168 ± 25 | 219 ± 16 |
| **Available-C:P ratio (*)** | **F=4.90 $P$=0.047** | $R^2$m=0.15 $R^2$c=0.86 | **$R^2$m=0.093 $R^2$c=0.72** | $P$=0.0035 | **362 ± 66b** | **2107 ± 367a** |
| Available-C:K ratio | F=2.67 $P$=0.12 | **$R^2$m=0.066 $R^2$c=0.59** | $R^2$m=0.057 $R^2$c=0.47 | $P$=0.76 | 20.6 ± 3.1 | 34.2 ± 3.6 |
| Available-N:P ratio | F=3.0 $P$=0.097 | **$R^2$m=0.081 $R^2$c=0.67** | $R^2$m=0.081 $R^2$c=0.67 | $P$=0.99 | 2.72 ± 0.45 | 10.6 ± 1.8 |
| Available-N:K ratio | F=0.17 $P$=0.68 | **$R^2$m=0.0049 $R^2$c=0.59** | $R^2$m=0.0049 $R^2$c=0.59 | $P$=0.99 | 0.158 ± 0.024 | 0.177 ± 0.017 |
| Available-P:K ratio | F=3.54 $P$=0.072 | $R^2$m=0.11 $R^2$c=0.84 | **$R^2$m=0.08 $R^2$c=0.80** | $P$=0.012 | 0.104 ± 0.030 | 0.048 ± 0.008 |
| **Rate of $CO_2$ production (μg $g^{-1}$ $d^{-1}$) (*)** | **F=6.17 $P$=0.020** | **$R^2$m=0.17 $R^2$c=0.70** | $R^2$m=0.17 $R^2$c=0.70 | $P$=0.99 | **3.03 ± 0.22b** | **4.25 ± 0.17a** |
| Rate of $CH_4$ production (ng $g^{-1}$ $d^{-1}$) | F=3.57 $P$=0.071 | **$R^2$m=0.11 $R^2$c=0.87** | $R^2$m=0.11 $R^2$c=0.87 | $P$=0.99 | 5.02 ± 0.65 | 17.4 ± 2.6 |
| Rate of $N_2O$ production (ng $g^{-1}$ $d^{-1}$) | F=1.46 $P$=0.24 | $R^2$m=0.024 $R^2$c=0.16 | **$R^2$m=0.034 $R^2$c=0.091** | $P$=0.11 | 5.67 ± 1.52 | 3.96 ± 0.52 |
| Bulk density (g/$cm^3$) | F=0.250 $P$=0.62 | **$R^2$m=0.0093 $R^2$c=0.94** | $R^2$m=0.0093 $R^2$c=0.94 | $P$=0.99 | 0.873 ± 0.042 | 0.926 ± 0.019 |
| **pH (*)** | **F=8.69 $P$=0.0070** | **$R^2$m=0.24 $R^2$c=0.94** | $R^2$m=0.24 $R^2$c=0.93 | $P$=0.22 | **5.87 ± 0.14a** | **5.15 ± 0.07b** |
| Salinity (mS $cm^{-1}$) | F=2.0 $P$=0.23 | **$R^2$m=0.023 $R^2$c=0.14** | $R^2$m=0.008 $R^2$c=0.16 | $P$<0.0001 | 0.162 ± 0.016 | 0.110 ± 0.023 |
| Clay concentration (%) | F=0.063 $P$=0.80 | **$R^2$m=0.0023 $R^2$c=0.93** | $R^2$m=0.0024 $R^2$c=0.93 | $P$=0.40 | 10.3 ± 1.0 | 9.9 ± 0.5 |

| | | | | | | |
|---|---|---|---|---|---|---|
| Silt concentration (%) | F=1.95 *P*=0.18 | **$R^2$m=0.066 $R^2$c=0.91** | $R^2$m=0.066 $R^2$c=0.91 | *P*=0.99 | 44.8 ± 3.7 | 37.9 ± 1.1 |
| Sand concentration t (%) | F=1.42 *P*=0.25 | **$R^2$m=0.049 $R^2$c=0.92** | $R^2$m=0.049 $R^2$c=0.92 | *P*=0.99 | 44.9 ± 4.5 | 52.2 ± 1.4 |
| Air temperature (°C) | F=1.63 *P*=0.21 | **$R^2$m=0.060 $R^2$c=0.99** | $R^2$m=0.060 $R^2$c=0.99 | *P*=0.99 | 20.4 ± 0.2 | 19.5 ± 0.2 |
| **MAP (mm) (*)** | **F=14.6 *P*<0.0001** | **$R^2$m=0.36 $R^2$c=0.99** | **$R^2$m=0.36 $R^2$c=0.99** | *P*=0.98 | **1393 ± 19b** | **1617 ± 20a** |

## Figure Captions

**Fig. 1.** The location of the study area and sampling sites in Fujian Province, southeastern China.

**Fig. 2.** DFA comparing the groups of sites with low, medium and high rates of soil $CO_2$ production as a function of soil pH, available P by soil mass and available P by soil area as grouping factors.

**Fig. 3.** DFA comparing the groups of sites with low, medium and high rates of soil $CH_4$ production as a function of soil LOC concentration, LOC content, C concentration, N concentration and the N:K and C:K concentration ratios as grouping factors.

**Fig. 4.** DFA comparing the groups of sites with low, medium and high rates of soil $N_2O$ production as a function of soil air temperature, $Fe^{2+}$ content, $Fe^{2+}$ concentration and the LOC:N concentration ratio as grouping factors.

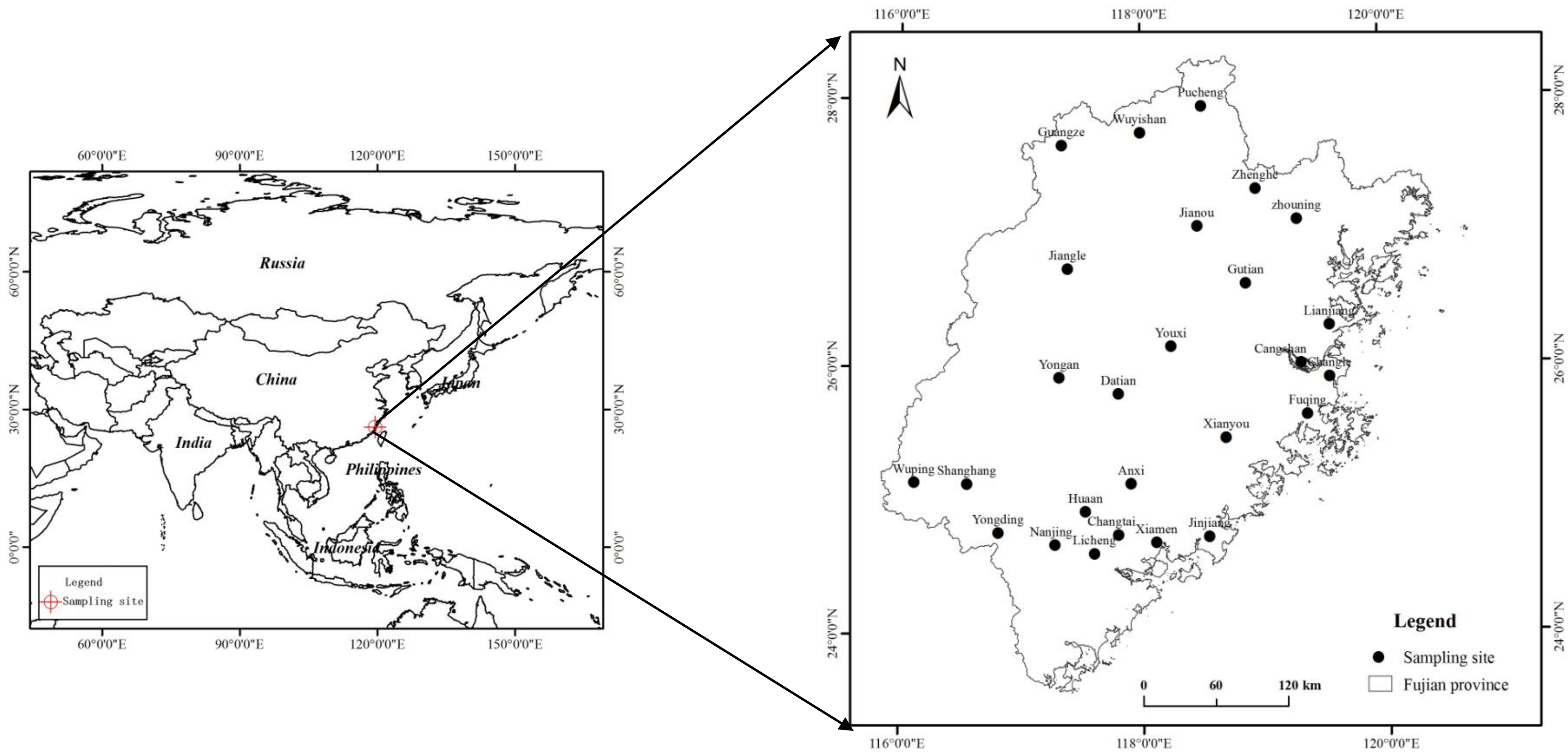

60°0'0"E
90°0'0"E
120°0'0"E
150°0'0"E
60°0'0"N
30°0'0"N
0°0'0"
Russia
China
India
Japan
Philippines
Indonesia
Legend
Sampling site
116°0'0"E
118°0'0"E
120°0'0"E
28°0'0"N
26°0'0"N
24°0'0"N
N
Pucheng
Wuyishan
Guangze
Zhenghe
zhouning
Jianou
Jiangle
Gutian
Lianjiang
Youxi
Cangshan
Changle
Yongan
Datian
Fuqing
Xianyou
Wuping
Shanghang
Anxi
Huaan
Yongding
Changtai
Xiamen
Jinjiang
Nanjing
Licheng
0
60
120 km
Legend
Sampling site
Fujian province

**Fig. 1**

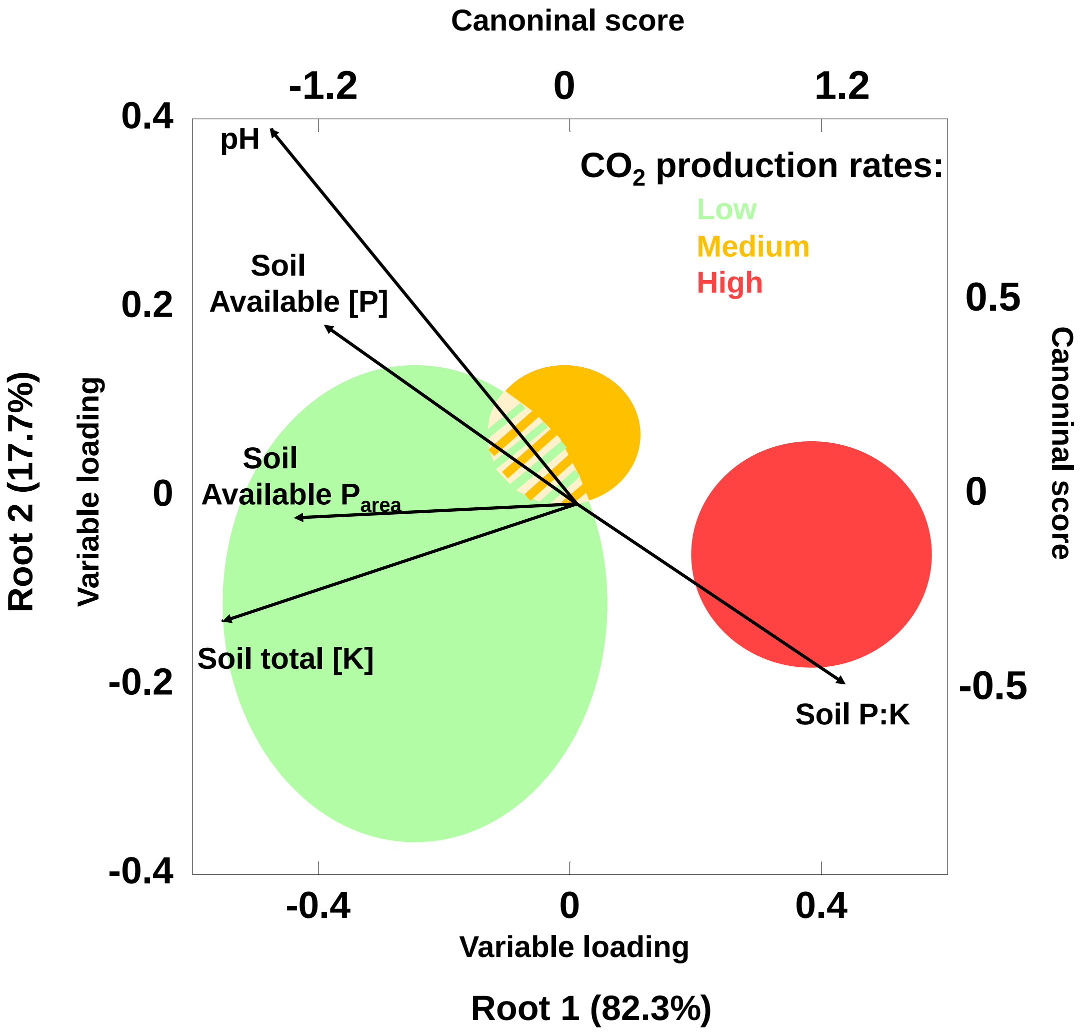

Canoninal score
-1.2
0
1.2
0.4
0.2
0
-0.2
-0.4
Root 2 (17.7%)
Variable loading
pH
Soil
Available [P]
Soil
Available $P_{area}$
Soil total [K]
Soil P:K
$CO_2$ production rates:
Low
Medium
High
0.5
0
-0.5
Canoninal score
-0.4
0
0.4
Variable loading
Root 1 (82.3%)

**Fig. 2**

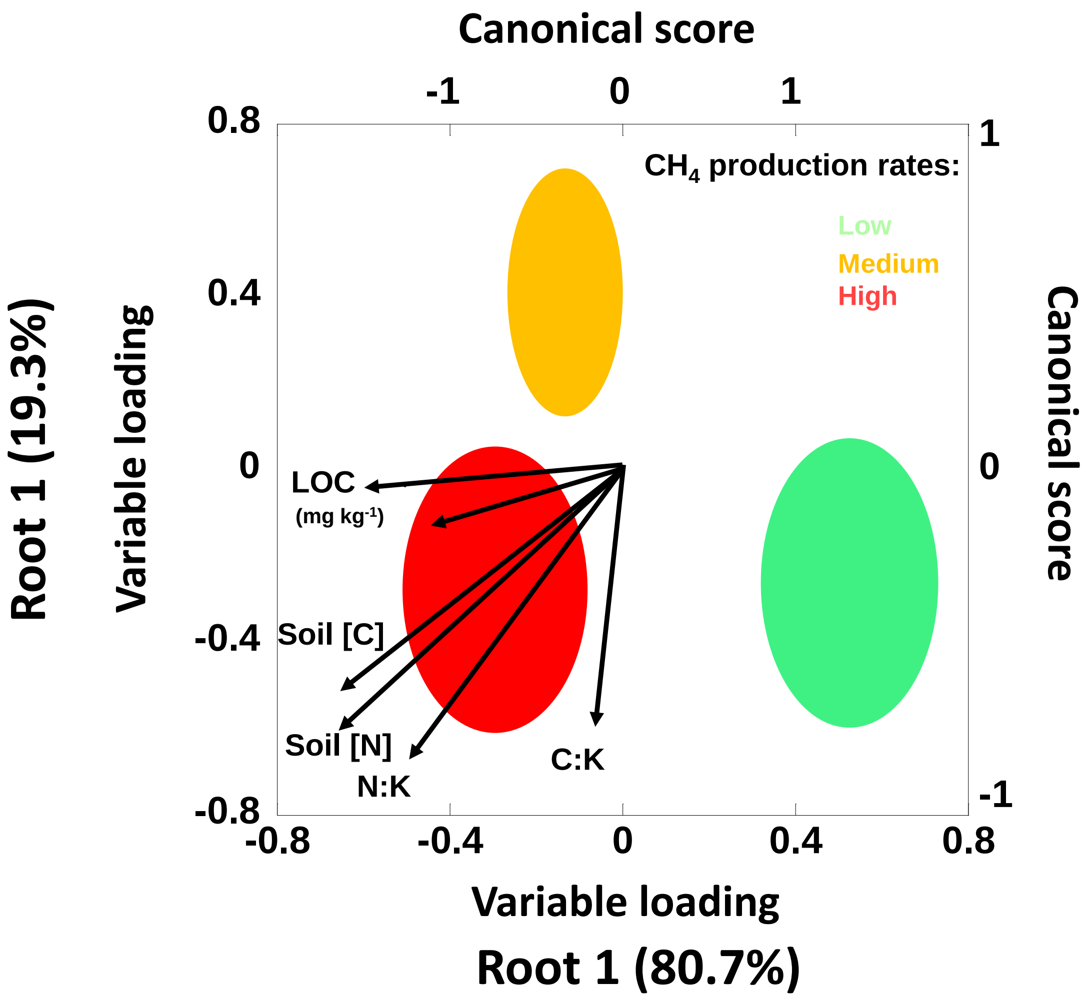
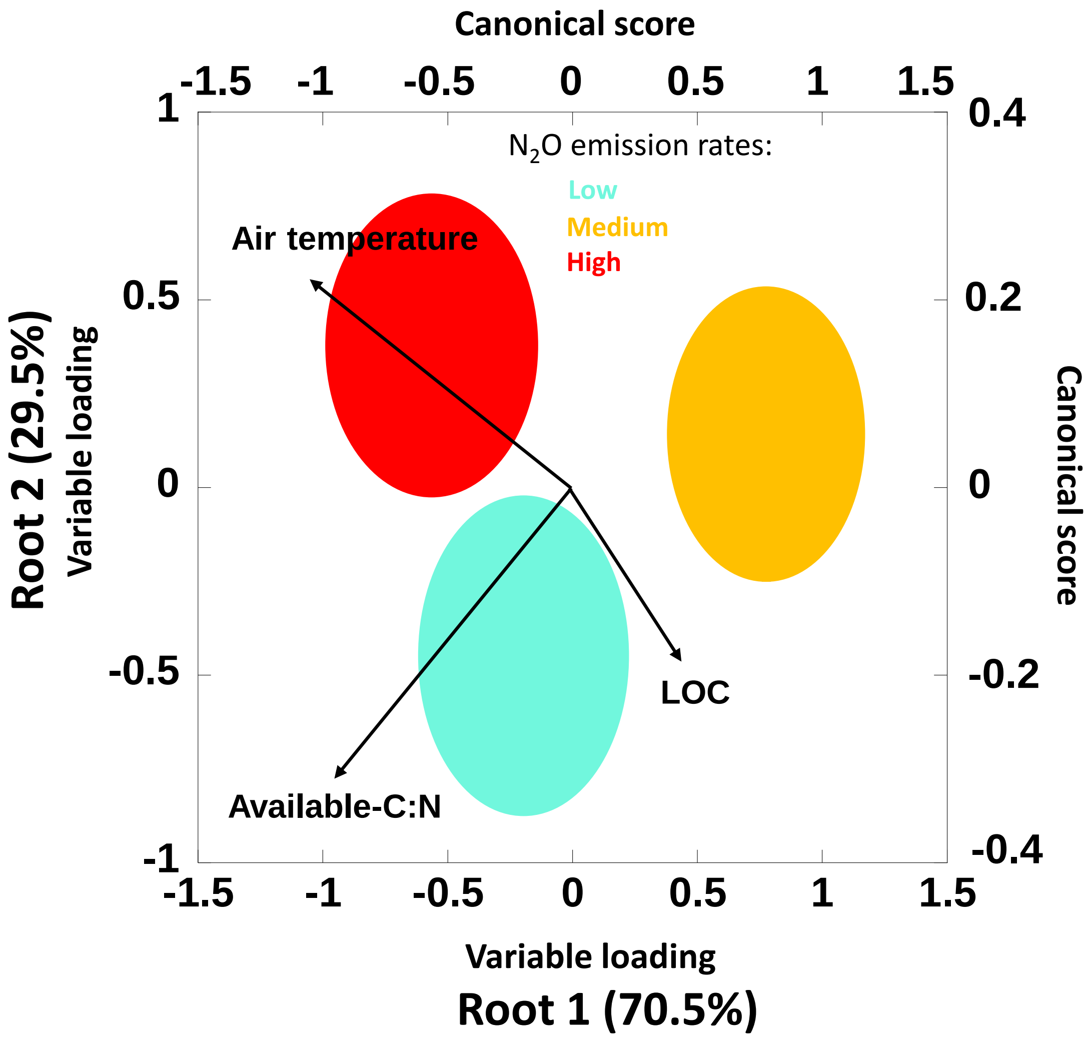

Canonical score
-1
0
1
0.8
0.4
0
-0.4
-0.8
1
0
-1
CH4 production rates:
Low
Medium
High
LOC
(mg kg-1)
Soil [C]
Soil [N]
N:K
C:K
-0.8
-0.4
0
0.4
0.8
Variable loading
Root 1 (80.7%)
Root 1 (19.3%)
Variable loading
Canonical score

**Fig. 3**



Canonical score
-1.5
-1
-0.5
0
0.5
1
1.5
N2O emission rates:
Low
Medium
High
Air temperature
LOC
Available-C:N
Root 2 (29.5%)
Variable loading
Canonical score
0.4
0.2
0
-0.2
-0.4
1
0.5
0
-0.5
-1
Variable loading
Root 1 (70.5%)


**Fig. 4**

Supplementary Material
Click here to download Supplementary Material: Supplementary material.docx