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## Stand age amplifies greenhouse gas and NO releases following conversion of rice paddy to tea plantations in subtropical China



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#### ABSTRACT

Increasing demand of commercial markets for tea products is driving the conversion from rice paddies to tea plantations in subtropical China. So far, however, little is known on how this land-use change, along with the age of plantation establishment, will affect the fluxes of greenhouse gases (GHG) methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) and the air-pollutant nitric oxide (NO). Thus, we measured CH<sub>4</sub>, N<sub>2</sub>O and NO fluxes over an entire year from two tea plantations (one- and five-year-old plantations) and a native rice-fallow system under two systemspecific nitrogen fertilizer options. On an annual scale, comparable or higher GHG emissions were measured for tea plantations as compared to the rice-fallow system. Besides the pollution swapping from  $CH_4$  being the dominating GHG in rice paddies to N<sub>2</sub>O in tea plantations, annual NO emissions increased significantly. Annual direct emission factors of N<sub>2</sub>O and NO in tea plantations were 2.47-5.80% and 2.00-3.99%, respectively, significantly higher than in the rice-fallow system (N<sub>2</sub>O: 1.05–2.05%; NO: 0.033–0.051%). Differences in N fertilizer inputs and soil environmental conditions (e.g., soil water regime, pH and organic carbon) due to contrasting managements of these systems and their interactions are clearly driving the stimulation of N2O and NO emissions and contributing to the significant CH<sub>4</sub> reductions. Furthermore, we observed that increasing tea stand age, particularly under organic fertilization, further stimulated N<sub>2</sub>O and NO emissions that varied significantly intraannually. Nevertheless, the higher N<sub>2</sub>O and NO emissions of tea plantations and the increasing emission strength with stand age deserve further research attention and consideration for future land-use conversions.

#### 1. Introduction

Anthropogenic emissions of greenhouse gases (GHG) are major drivers of global climate change (Montzka et al., 2011), with methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) contributing approximately 17% and 6% to the total global increase in radiative forcing, respectively (WMO, 2014). Moreover, anthropogenic nitric oxide (NO) emissions are considered as the main precursor of ground-level tropospheric ozone (O<sub>3</sub>), impacting human health and ecosystem functioning (Ludwig et al., 2001; Medinets et al., 2015). There is compelling evidence that agriculture, and here specifically fertilized agricultural soils, are key sources of N<sub>2</sub>O and NO as well as for CH<sub>4</sub>. In this context, rice cultivation, with waterlogged soils supporting methanogenesis, is a significant source of global CH<sub>4</sub> emissions, accounting for about 10% of all anthropogenic CH<sub>4</sub> emissions (Nazaries et al., 2013). Although drained or well-aerated upland systems can be a net sink of atmospheric CH<sub>4</sub>, emissions of N<sub>2</sub>O and NO from the application of nitrogenous fertilizers in these systems were estimated to be 4.1 Tg N<sub>2</sub>O-N yr<sup>-1</sup> and 3.7 Tg NO-N yr<sup>-1</sup>, respectively, contributing approximately 60% and 10%, respectively, to the total global anthropogenic emissions (Intergovernmental Panel on Climate Change IPCC, 2013). Indeed, land-use practices, which are regarded as the second largest anthropogenic source of GHG emissions (Intergovernmental Panel on Climate Change IPCC, 2013), can have strong influences on the C- and N-trace gas emissions because of management-induced changes in crop types and in soil physical, chemical and microbial properties (Zhao et al.,

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2016; Weller et al., 2016). Accordingly, it is critical to assess the impacts of land-use changes on GHG and NO emissions and understand the underlying mechanisms, which is of great importance for improving our knowledge on these emissions and realizing the potential of different managements on climate change mitigation.

Globally, China is one of the most important rice-producing countries, contributing about 35% to the total global rice production (Qu et al., 2012). Numerous studies, including field measurements and model simulations, have focused on CH<sub>4</sub> and N<sub>2</sub>O emissions from Chinese rice cultivations, and annual total emissions of CH<sub>4</sub> and N<sub>2</sub>O from China's rice systems have been estimated as  $6-10 \text{ Tg CH}_4 \text{ yr}^{-1}$  and 32–51 Gg N<sub>2</sub>O-N yr<sup>-1</sup>, respectively (Zou et al., 2009; Yan et al., 2009; Liu et al., 2016). Yet, few studies have measured NO fluxes from Chinese rice cultivations (Yan et al., 2003; Zhou et al., 2010). Rice is a profligate user of water, i.e., producing 1 kg of rice needs approximately 3000-5000 Liters of water, significantly higher than the water footprints of upland crops (Bouman et al., 2002). For the terraced rice fields in hilly regions of subtropical China, however, irrigation remains challenging due to the imperfect infrastructure. One option for local farmers to adapt to this situation is to convert traditional rice-fallow cropping systems into tea plantations. Moreover, recent socioeconomic developments with increased market demands for tea products are becoming the most important impulse for rice farmers cultivating tea plants in order to gain higher economic returns. It is estimated that China's total area of tea plantation has reached 1.85 million ha in 2009, accounting for 52% of the world total (Han et al., 2013), whereas the area under rice cultivation shows a decreasing trend in recent decades (Sun et al., 2011). In contrast to rice, tea is a leaf-harvested crop, and nitrogen is the most important essential nutrient for increasing the content of free amino acids, the major quality indicator of tea leaves (Oh et al., 2006). To obtain high quality tea leaves, a large amount of inorganic and organic nitrogen fertilizers are increasingly applied in tea plantations. The annual application rate of nitrogen fertilizer in tea plantations is as high as  $450-1200 \text{ kg N} \text{ ha}^{-1} \text{ yr}^{-1}$  (Tokuda and Hayatsu, 2004; Hirono and Nonaka, 2012; Yao et al., 2015), which is much higher than the fertilizer application rates in rice cultivations which are in a range of 100–360 kg N ha<sup>-1</sup> for a crop season (Zou et al., 2009). Not surprisingly, such high nitrogen application in tea plantations without compensatory soil liming results in significant soil acidification (Han et al., 2007) and high potential rates of environmentally harmful N-containing trace gases (N2O and NO) release (Yao et al., 2015). Accordingly, Akiyama et al. (2006) and Han et al. (2013) found that N<sub>2</sub>O emissions from tea plantations might be much higher relative to other upland fields or rice paddies.

Land conversion from rice cultivation to tea plantation means that a predominantly flooded system, with soils being water saturated and anaerobic in most of the year, is turned to an upland system. As has been shown for management changes from a rice-based system to maize-based systems, this might lead to pollution swapping, i.e., decreased emissions of CH4 at the expense of increases in N2O emissions (Stevens and Quinton, 2009; Weller et al., 2016). A study conducted under subtropical climate conditions in China reported that the conversion of rice paddies to aquaculture significantly reduced CH<sub>4</sub> and N<sub>2</sub>O emissions by 48% and 56%, respectively (Liu et al., 2016). In contrast, the conversion of rice paddies to fertilized vegetable fields in the same climate regions showed no significant impact on total GHG (CH<sub>4</sub> and N<sub>2</sub>O) emissions (Yuan et al., 2016). Nevertheless, so far there has been very limited data available on GHG and NO emissions from tea plantations in subtropical China (Li et al., 2013; Han et al., 2013; Yao et al., 2015; Fu et al., 2015). Moreover, tea plantations with different ages of establishment represent a practical system in subtropical China. Due to high N fertilization rates in combination with inherently physiological feature of tea plants and its litter, soil environmental properties such as specifically soil pH are changing with the age of a tea plantation. Several studies have shown that these changes are affecting C and N availability, gross N transformation rates, and microbial

biomass, activity and community composition (Xue et al., 2006; Han et al., 2007; Kamau et al., 2008; Zhu et al., 2014). However, whether tea stand age also affects GHG and NO emissions is still unknown.

In our study we hypothesized that conversion of rice paddies to tea plantations will lead to a significant pollution swapping from a CH<sub>4</sub> dominated GHG balance to a N<sub>2</sub>O dominated GHG balance, while the GHG emission strength remains comparable for both systems. Moreover, we hypothesized that tea stand age matters and that N<sub>2</sub>O as well as NO emissions increase with stand age. With increasing age of tea plantation soil organic C content increases, while due to extensive Nfertilizer application without compensatory lime addition soil pH decreases: both factors likely promoted N2O and NO productions by microbial processes as well as by chemo-denitrification. We furthermore hypothesized that fertilizer management has a significant impact on Cand N-trace gas emissions and that the use of organic fertilizers in tea plantations might reduce N-trace gas emissions, while split versus single application of N fertilizer for rice production will result in higher CH<sub>4</sub> emissions due to improved crop growth and higher rice root C exudation.

To address these hypotheses, we performed field measurements of  $CH_4$ ,  $N_2O$  and NO fluxes over an entire year from two tea plantations with different stand ages (1yr and 5 yr) and compared fluxes to those of adjacent rice cultivations under two system-specific nitrogen fertilizer application options. For the rice-fallow system, the evaluated N fertilizer practices were single and split applications; while for tea plantations synthetic N fertilizer (urea) and organic fertilizer (oilcake) were tested.

#### 2. Materials and methods

#### 2.1. Site description, experimental design and crop management

The field study was conducted at a farm  $(32^{\circ}07' \text{ N}, 110^{\circ}43' \text{ E}; 440 \text{ m}$ above sea level) of the Agricultural Bureau of Fangxian County in China, from September 2013 to September 2014. The region is characterized by a northern subtropical monsoon climate, with a mean annual air temperature of 14.2 °C and a mean annual precipitation of 914 mm (Yao et al., 2015). In this area, the dominating cropping systems are paddy rice-winter fallow or paddy rice-oilseed rape rotations. However, in recent years tea plantations were increasingly established on former rice paddy fields.

In this study, we investigated two tea plantations (T) and one adjacent paddy rice-fallow cropping system (P). Experiments were arranged in a split-block design with quadruplicate fields for each cropping system. The two tea plantations were constructed from paddy ricefallow cropping systems in 2012 and 2008, and were one- (T1) and fiveyear-old (T5), respectively, at the start of experiments. Before tea plantations were established, all experimental fields were farmed as rice-based cropping systems under the same management regimes for decades, so that soil homogeneity across all treatments can be assumed. Following the establishment of tea plantations, soils were under regular fertilization of 450 kg N ha<sup>-1</sup> yr<sup>-1</sup> with synthetic nitrogen fertilizers or organic fertilizers following common regional management practices. The main soil physicochemical properties of each cropping system were determined in September 2013 (12 soil samples were taken from 0 to 15 cm depth in each system) and are shown in Table 1. Two fertilizer treatments for rice and tea systems were further established, reflecting commonly used fertilization practices by farmers (Table S1). In the tea system (T1 and T5), as determined by farm survey, urea (UN) or oilcake (OM) was applied at an annual rate of  $450 \text{ kg N} \text{ ha}^{-1} \text{ yr}^{-1}$ , with fertilizer being applied in two splits (i.e., 150 and 300 kg N ha<sup>-1</sup> in the autumn and spring seasons, respectively) in the inter-rows using the band application method (Yao et al., 2015). In the rice system (P), in agreement with recommended N application rates for this region (Zhu and Chen, 2002), urea was applied as a single dose before rice transplanting at a common rate of 150 kg N ha<sup>-1</sup> (UN) or in split application

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