Effects of land use conversion and fertilization on CH₄ and N₂O fluxes from typical hilly red soil

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Abstract Land use conversion and fertilization have been widely reported to be important managements affecting the exchanges of greenhouse gases between soil and atmosphere. For comprehensive assessment of methane (CH₄) and nitrous oxide (N₂O) fluxes from hilly red soil induced by land use conversion and fertilization, a 14-month continuous field measurement was conducted on the newly converted citrus orchard plots with fertilization (OF) and without fertilization (ONF) and the conventional paddy plots with fertilization (PF) and without fertilization (PNF). Our results showed that land use conversion from paddy to orchard reduced the CH₄ fluxes at the expense of increasing the N₂O fluxes. Furthermore, fertilization significantly decreased the CH₄ fluxes from paddy soils in the second stage after conversion, but it failed to affect the CH₄ fluxes from orchard soils, whereas fertilizer applied to orchard and paddy increased soil N₂O emissions by 68 and 113.9 %, respectively. Thus, cumulative CH₄ emissions from the OF were 100 % lower, and N₂O emissions were 421 % higher than those from the PF. Although cumulative N₂O emissions were stimulated in the newly converted orchard, the strong reduction of CH₄ led to

lower global warming potentials (GWPs) as compared to the paddy. Besides, fertilization in orchard increased GWPs but decreased GWPs of paddy soils. In addition, measurement of soil moisture, temperature, dissolved carbon contents (DOCs), and ammonia (NH₄⁺-N) and nitrate (NO₃⁻-N) contents indicated a significant variation in soil properties and contributed to variations in soil CH₄ and N₂O fluxes. Results of this study suggest that land use conversion from paddy to orchard would benefit for reconciling greenhouse gas mitigation and citrus orchard cultivation would be a better agricultural system in the hilly red soils in terms of greenhouse gas emission. Moreover, selected fertilizer rate applied to paddy would lead to lower GWPs of CH₄ and N₂O. Nevertheless, more field measurements from newly converted orchard are highly needed to gain an insight into national and global accounting of CH₄ and N₂O emissions.

Keywords Land use conversion · Methane · Nitrous oxide · Fertilization · Global warming potentials

Introduction

The atmospheric trace gases methane (CH₄) and nitrous oxide (N₂O), two major potent long-lived greenhouse gases (GHGs), exhibit relative global warming potentials (GWPs) of 34 and 298 times that of CO₂ on mass basis over the 100-year time horizon, respectively (IPCC 2013). Approximately 70–90 % of the recent increases in CH₄ and N₂O concentrations in the atmosphere originate from biological sources, especially from agriculture ecosystems (Baruah et al. 2010). Paddy field, in particular, has been identified as one of the major sources of atmospheric CH₄ and N₂O. The periodic waterlogging-drainage alteration episodes and intensive fertilizer inputs in paddy field could provide suitable soil

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environment and accessible substrate for CH₄ and N₂O emissions (Hou et al. 2012; Yao et al. 2012). While paddy soils release substantial amounts of CH₄ and N₂O to the atmosphere, the changes in agriculture land use could have potentials for mitigating net global warming potential by decreasing CH₄ and/or N₂O emissions (Weller et al. 2016).

Land use change, which is regarded as the second largest anthropogenic source of greenhouse gas emissions (IPCC 2013), can substantially alter the dynamics of soil gases (Merino et al. 2004; Zona et al. 2013). However, results from current studies were not consistent, ranging from a large increase (Weller et al. 2016) to a decrease (Jiang et al. 2009) or to no significant impact (Inubushi et al. 2003) on soil CH₄ and N₂O fluxes. The large variability in the effect of land use change on soil CH₄ and N₂O fluxes was likely linked to specific site conditions, such as soil type, the type of land use change, and management practice used (Inubushi et al. 2003; Lin et al. 2012). Moreover, most of the existing studies have focused on the comparison of different types of land uses that have been converted for many years (Merino et al. 2004; Iqbal et al. 2009; Liu et al. 2011); little research has been done to understand the dynamics of CH₄ and N₂O fluxes and the mechanisms behind the observed changes during the initial stages after land use conversion.

China is one of the most important rice-producing countries in the world, accounting for 20 % of the world's rice production area and 23 % of all cultivated land in China (Zhang et al. 2010). The red soil, one of the typical agricultural soils in subtropical China, covers about 11.8 % of the country land surface and produces 80 % of the rice and supports 22.5 % of the population of China (Lou et al. 2003). During the past decades, the red soil regions, being the most densely populated, have been experiencing significant changes in land use. In particular, converting paddy fields to upland cultivation for growing vegetables, fruits, and economic forest has been locally advocated to meet its increasing market demands and gain higher economic returns in these regions (Lin et al. 2012). Such conversions could not only alter the physical, chemical, and biological properties of the soil but also significantly impact the GHG emissions. However, there are only few reports on the soil CH₄ and N₂O fluxes, and their relationships with management practices have been available during land use conversion in the red soil regions of China, especially referring to the conversion from paddy field to upland. In this study, a 14-month field measurement of CH₄ and N₂O fluxes was simultaneously taken from conventional paddy fields and the newly paddy field-converted citrus plantation in a typical hilly red soil region of southern China. The main objectives of this study were to (1) gain an insight into the dynamics of CH₄ and N₂O fluxes during land use conversion from paddy field to citrus plantation, (2) assess the impact of environmental factors and fertilization on soil CH₄ and N₂O fluxes, and (3) examine whether the conversion of rice paddy

to citrus plantation could have a potential for mitigating the CH₄ and N₂O emissions. Such information is expected to be helpful in developing appropriate management practices for mitigating climatic impacts in this region.

Materials and methods

Site description

The experimental site was located at the Qianyanzhou Experimental Station (26° 44' N, 115° 04' E) of the Chinese Academy of Science in Jiangxi province, southern China. The site is characterized by a subtropical monsoon climate with a mean annual air temperature of 18.0 °C and a mean annual precipitation of approximately 1509.0 mm during 1989–2010. The soil was the typical red soil found in middle subtropical China and classified as Cambosols (Ultisols classification), which were derived from sandstone and sandy conglomerate. Double cropping of paddy is the main cropping systems in this region, with late rice (late July to late November), fallow period (late November to next late April), and early rice (late April to late July) in rotation. Soil texture was sandy loam with 58 % sand, 31 % silt, and 11 % clay. The topsoil (0–10 cm) has an organic carbon content of 9.40 g kg⁻¹ and total N content of 1.00 g kg⁻¹. The soil pH was 4.99, and the bulk density was 1.30 g cm⁻³ (Yuan et al. 2015).

Experimental design

The two most prevalent uses of agricultural land in southern China were selected for this study: rice paddy (*Oryza sativa* L.) and citrus orchard (*Citrus reticulata*). The study area was previously paddy field and had been successively converted to orchard in June 2012. To evaluate the effects of fertilization, fertilizer was added according to the local fertilizer rate (Dong et al. 2012; Lin et al. 2012). In addition, a non-fertilization treatment was also established. Thus, four treatments were established, namely citrus orchard with fertilization (OF) and without fertilization (ONF) and rice paddy with fertilization (PF) and without fertilization (PNF). All treatments were arranged in a randomized block design with four replicates, for a total of 16 experimental plots (with a size of 12 × 14 m), which were separated by buffer strips. According to the local management, the basal fertilizers were amended to a depth of 50 cm before the orchard established to ensure the survival of the trees. In the PF, compound fertilizer was broadcasted during rice transplanting and urea was broadcasted in the form of top dressings at the tillering stage, whereas in the OF, compound fertilizer combined with urea was uniformly spread on the soil surface. Besides, in the paddy fields, irrigation was manually supplied during the early days following rice transplantation in the conventional cultivation in order to ensure

survival and yield. The details of the cultivation and fertilization schemes are shown in Table 1.

Measurement of soil CH₄ and N₂O fluxes

The soil CH₄ and N₂O fluxes were measured using static opaque chambers and gas chromatography techniques from June 2012 to July 2013. Stainless steel collars (covering an area of 0.12 m²) were inserted into the soil to a depth of 0.1 m. Chambers with a bottom area of 0.12 m² and height of 0.39 or 0.69 m (according to the plant height) were temporarily mounted onto the frames for gas sampling. The edges of the chambers and lids were sealed with rubber to prevent leakage. Besides, the chambers were covered with thermal insulation to reduce heating from direct solar radiation during sampling. Then, chambers were mounted onto the installed base frames during sampling period. In addition, the base frames were kept in the same location throughout the whole study period in the orchard, whereas those in the paddy fields were removed before tillage and were placed (24 h before the measurement) in the location marked for subsequent measurements.

Trace gas fluxes were measured daily for 5 days after fertilization and once or twice a week for the remaining period. Gas samplings were performed between 09:00–11:00 AM local time. Five air samples were withdrawn at an interval of 10 min using 100-ml plastic syringes attached to a three-way stopcock closure and then injected into evacuated bags made of inert aluminum-coated plastic. Within 24 h following gas collection, the gas samples were analyzed with a gas chromatography (Agilent 7890A) equipped with a flame ionization detector for CH₄ detection at 200 °C, an electron capture detector for N₂O detection at 330 °C, and the temperature for column was set at 55 °C. The fluxes were calculated from the linear changes in gas concentrations in the enclosed chamber headspace with time. During gas sampling, the temperature of the chamber was recorded using a portable digital

thermometer and then used in the calculation of the actual gas concentration changes.

Auxiliary measurements

Daily precipitation and air temperature were obtained from the Qianyanzhou meteorological station. Soil temperature and water content were recorded every 10 min by an automatic data logging system through wireless technology, with temperature and humidity probes being buried in each plot. In addition, the floodwater depths in the paddy were measured daily using a ruler in the flooding period. Surface soil (0–10 cm) samples were collected twice or three times a month between June 2012 and July 2013. For ammonia (NH₄⁺-N) and nitrate (NO₃⁻-N) analysis, 20 g of fresh soil was extracted with 100 ml of a potassium chloride solution (1 M, shaken for 1 h) and filtered with filter paper, and subsequently, extracts were analyzed with a continuous flow auto analyzer (Seal AA3, Germany). Soil dissolved carbon (DOC) was extracted by adding 100 ml of distilled water to 20 g of fresh soil, shaking this mixture for 1 h, and filtering it through fiber filters and then analyzed with a Total Organic Carbon analyzer (Elementar vario TOC, Germany).

Statistical analyses

To assess the effects of the basal fertilization in the orchard plots, we divided the observing period into the first (from August 2012 to January 2013) and second (from February 2013 to July 2013) stage after conversion. Cumulative GHG emissions were directly computed from the measured fluxes and were estimated by linear interpolation for days until no measurements were made. Using the IPCC (IPCC 2013) standard for calculating the combined GWPs (CO₂ eq) of CH₄ and N₂O for 100 years that

$$\text{CO}_2 \text{ eq} = 34 \times \text{CH}_4 + 298 \times \text{N}_2\text{O} \text{ kg CO}_2 \text{ equivalent ha}^{-1}$$

Table 1 Management practices for the experimental fields

Fields	Management	Date
Orchard	Draining and rotary tillage	June 20, 2012
	Orchard planting, and basal fertilizer (370 kg N ha ⁻¹) was applied to the fertilized fields	July 30, 2012
	Compound fertilizer (67.2 kg N ha ⁻¹) and urea (20.8 kg N ha ⁻¹) were applied to the fertilized fields	March 26, 2013
	Compound fertilizer (67.2 kg N ha ⁻¹) and urea (20.8 kg N ha ⁻¹) were applied to the fertilized fields	June 17, 2013
Paddy	Rice transplanting, and compound fertilizer (72 kg N ha ⁻¹) was applied to the fertilized fields	July 30, 2012
	Urea (106.2 kg N ha ⁻¹) was applied to the fertilized fields	August 10, 2012
	Rice harvest	November 14, 2012
	Rice transplanting, and compound fertilizer (72 kg N ha ⁻¹) was applied to the fertilized fields	April 24, 2013
	Urea (106.2 kg N ha ⁻¹) was applied to the fertilized fields	May 3, 2013
	Rice harvest	July 22, 2013

Repeated measures of one-way analysis of variance (ANOVA) were conducted to test the differences in soil properties and CH_4 and N_2O fluxes in all stages after land use conversion among the four treatments. ANOVA was performed to evaluate the difference of cumulative CH_4 and N_2O emissions. A general linear model for univariate ANOVA was used to evaluate the significant differences in land use conversion, fertilization, and their interactions. Pearson correlation coefficients and multiple regression analysis were used to examine the relationships between $\text{CH}_4/\text{N}_2\text{O}$ fluxes and soil properties. All statistical analyses were conducted using SPSS version 16.0 (SPSS Inc., USA).

Results

Environmental and soil properties

The maximum and minimum daily air temperatures during our observing period were 31.6°C (July 2012) and -0.1°C (January 2013), respectively, with a mean air temperature of 18.6°C (Fig. 1a). The annual precipitation at the experimental site was 1321.8 mm, lower than the long-term average (1506.4 mm) and monthly precipitation that ranged from 30.4 to 196.0 mm (Fig. 1a). Floodwater depths in the paddy

were above the soil surface in the early stage of paddy growing (Fig. 1b), which would lead to a waterlogging period. The variations of soil water content in orchard plots were related to the precipitation (Fig. 1b), but were not affected by fertilization (Table 2). Soil temperature at 5-cm depth fluctuated greatly, which correlated with the weather conditions (Fig. 1c). Land use conversion clearly increased soil temperature, whereas fertilization did not affect it (Table 2).

The $\text{NH}_4^+\text{-N}$ ranged from 3.26 to 24.08 and from 3.12 to 17.30 mg kg^{-1} (Fig. 2a), while the $\text{NO}_3^-\text{-N}$ ranged from 0.51 to 11.46 and from 0.21 to 9.55 mg kg^{-1} in the orchard and paddy soils, respectively (Fig. 2b). Land use conversion from paddy to orchard significantly increased the soil $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ concentration, with them in OF being 0.34 and 2.69 times, respectively, higher than those in PF ($P < 0.05$, Table 2). Fertilization significantly increased $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ concentrations throughout the study period ($P < 0.05$, Table 2). Furthermore, the soil DOC content was significantly lower in the orchard plots ($5.79\text{--}92.69 \text{ mg l}^{-1}$), when compared with that in the paddy plots ($4.66\text{--}217.81 \text{ mg l}^{-1}$) (Fig. 2c). Thus, conversion of the PF treatment into the OF treatment decreased mean DOC concentration from 78.22 to 29.58 mg l^{-1} ($P < 0.05$, Table 2). DOC concentrations in the OF and PF were 1.32 and 1.09 times higher than those in the ONF and PNF, respectively ($P < 0.05$, Table 2).

Fig. 1 Seasonal variation of daily precipitation and air temperature (a), floodwater depth in paddy fields and soil water content in orchard fields (b), and soil temperature (c) in the four treatments from June 2012 to July 2013. The data are shown as means

Soil CH_4 and N_2O fluxes

Average soil CH_4 and N_2O fluxes did not differ significantly in the four treatments prior to the conversion (Table 3). After land use conversion, the patterns of CH_4 fluxes significantly varied between the converted orchards and paddy fields (Fig. 3). In the newly converted orchard, short lasting strong CH_4 emissions at the early days after conversion and then negative CH_4 fluxes were observed (Fig. 3a). While the CH_4 fluxes from paddy soils peaked at around 20 days after transplanting of paddy cultivations and then sharply declined and became stabilized before harvesting (Fig. 3b). Compared to PF, the soil CH_4 fluxes from the OF and ONF were significantly lower in both stages after conversion (Table 3). While average CH_4 fluxes from orchard soils were positive values in the first stage, they were negative values in the second stage (Table 3). Fertilization could significantly decrease the CH_4 fluxes from paddy soils in the second stage, whereas there were no significant differences in CH_4 fluxes between OF and ONF (Table 3).

After land use conversion, soil N_2O fluxes gradually began to keep at positive values, and negative value was scarcely observed (Fig. 4a), whereas in paddy soils, negative values were observed especially in the waterlogging period (Fig. 4b). Fertilizer application triggered the flux peaks of

Table 2 Effects of land use conversion and fertilization on selected environment factors

Treatments	Soil temp (°C)	Soil water content (%)	NH ₄ ⁺ -N (mg kg ⁻¹)	NO ₃ ⁻ -N (mg kg ⁻¹)	DOC (mg l ⁻¹)
OF	21.00 ± 0.20a	36.82 ± 0.22a	12.58 ± 0.17a	4.32 ± 0.15a	29.58 ± 0.55c
ONF	20.55 ± 0.034a	37.60 ± 0.17a	8.23 ± 0.18c	3.23 ± 0.22b	22.40 ± 0.56d
PF	19.97 ± 0.12b	—	9.34 ± 0.31b	1.17 ± 0.058c	78.22 ± 2.81a
PNF	19.93 ± 0.16b	—	7.42 ± 0.16c	0.96 ± 0.039c	71.54 ± 0.69b
Significance of					
L	0.030	—	0.000	0.000	0.000
F	0.850	—	0.000	0.000	0.030
L × F	0.580	—	0.000	0.000	0.916

The data are shown as means with standard errors. Different letters in the same column indicate significant differences ($P < 0.05$) between corresponding treatments. L represents land use conversion; F represents fertilization; L × F represents interactions between land use conversion and fertilization. Significant effects ($P < 0.05$) are highlighted in bold

N₂O in both orchard and paddy fields in both stages after conversion (Fig. 4a, b). Thus, land use conversion and fertilization significantly increased average soil N₂O fluxes, following the order of OF > ONF > PF > PNF in both stages after land use conversion (Table 3).

Fig. 2 Seasonal variation of the soil NH₄⁺-N (a), NO₃⁻-N (b), and DOC (c) concentrations in the four treatments. The data are shown as means with standard errors. The black arrow indicates the cultivation of orchard and paddy. The gray arrows indicate fertilizer applied to PF, and the light gray arrows indicate fertilizer applied to OF. The legends and arrows in panel c applied to panels a and b as well

Cumulative emissions and GWPs of CH₄ and N₂O

The cumulative CH₄ and N₂O emissions over the observing period were dramatically influenced by land use conversion and fertilization (Fig. 5 and Table 4). Basing on the cumulative emissions 1 year after conversion, land use conversion significantly decreased soil CH₄ emissions ($P < 0.05$, Table 4). Annual CH₄ emissions from the OF and ONF treatment were approximately 100 and 100 % respectively, lower as compared to those from the PF treatment (Table 4). The CH₄ emissions from the PF were 124 % times lower than those from the PNF, whereas the cumulative CH₄ emissions for the OF were 4 % lower than those for the ONF (Table 4). Over the observing period, these N₂O emissions in the OF and ONF were 421 and 100 % times, respectively, greater than those in the PF. In contrast to the inhibitory effect of fertilization on the CH₄ emissions, the fertilizers applied to orchard and paddy induced 114 and 68 % increase in N₂O emissions, respectively (Table 4).

As shown in Table 4, the magnitude of CH₄ and N₂O emissions, expressed as CO₂ equivalents, differed significantly among treatments. Based on the summed annual emissions of both gases, land use conversion from paddy to orchard significantly reduced CO₂ eq of by 58 and 75 %, respectively (Table 4). Compared to CO₂ eq of the ONF treatment, this value for the OF was 69 % times higher (Table 4). However, fertilization for paddy showed no significant impact on soil CO₂ eq of CH₄ and N₂O emissions, though a 7 % reduction was observed from PF than those from PNF (Table 4).

Correlations between soil CH₄ and N₂O fluxes and soil properties

In contrast with a poor relationship between CH₄ fluxes and soil temperature in orchard fields, significant positive relationships between CH₄ fluxes and soil temperature

Table 3 Effects of land use conversion and fertilization on soil CH₄ and N₂O fluxes in different stages

	CH ₄ (μg C m ⁻² h ⁻¹)			N ₂ O (μg N m ⁻² h ⁻¹)		
Treatments	T0	T1	T2	T0	T1	T2
OF	462.10 ± 126.53a	15.30 ± 4.73b	−18.04 ± 1.73c	29.80 ± 3.81a	161.27 ± 12.14a	248.29 ± 22.21a
ONF	642.33 ± 89.03a	20.88 ± 8.39b	−24.17 ± 1.45c	30.89 ± 8.48a	139.35 ± 7.45a	99.33 ± 10.19b
PF	674.84 ± 136.80a	10,194 ± 2685.6a	3758.1 ± 791.32b	28.31 ± 2.30a	12.01 ± 0.55b	63.44 ± 7.08c
PNF	495.98 ± 124.90a	8046.9 ± 1149.3a	6183.9 ± 714.33a	33.68 ± 3.01a	7.73 ± 0.52c	26.06 ± 4.60d
Significance of						
L	—	0.000	0.000	—	0.000	0.000
F	—	0.131	0.000	—	0.092	0.003
L × F	—	0.294	0.000	—	0.090	0.003

The data are shown as means with standard errors. Different letters in the same column indicate significant differences ($P < 0.05$) between corresponding treatments. T0 means days before conversion; T1 means the first stage after conversion; T2 means the second stage after conversion. Significant effects ($P < 0.05$) are highlighted in bold

were found for paddy fields (Table 5). On the other hand, soil water content could positively affect soil CH₄ fluxes from orchard soils (Table 5). The dependence of CH₄ fluxes on soil DOC concentrations was observed irrespective of the land uses (Table 5). In orchard field, N₂O fluxes were positively related to soil temperature but negatively related to soil water content (Table 5), whereas in paddy plots, no significant relationships between N₂O fluxes and soil temperature were observed. There were significantly positive correlations between soil N₂O fluxes and NO₃[−] concentrations in both land uses ($P < 0.001$, Table 5). The results of multiple regression analysis confirmed the linear correlation between CH₄ fluxes and DOC concentration in the paddy and orchard plots, with R^2 of 77 and 74 %, respectively (Table 6), whereas the NO₃[−] contents accounted for 73 and 74 % of soil N₂O flux variability (Table 6).

Fig. 3 Seasonal variation of the methane (CH₄) fluxes from orchard (a) and paddy (b) soils. The data are shown as means with standard errors. The black arrow indicates the cultivation of orchard and paddy. The light gray arrows in panel a indicate fertilizer applied to OF, and the gray arrows in panel b indicate fertilizer applied to PF

Discussion

Comparison with other studies

In the past decades, significant lower CH₄ emissions but higher N₂O emissions were usually observed from upland soils as compared to paddy soils (Jiang et al. 2009; Weller et al. 2016). However, most of the previous studies emphasized the comparison of CH₄ and N₂O fluxes in different land use types and did not consider the process of land use conversion (Jiang et al. 2009; Lin et al. 2012; Zona et al. 2013). To our knowledge, this is the first in situ measurement of the CH₄ and N₂O fluxes from red soils in the early stage after land use conversion in mid-subtropical China. In the present study, annual CH₄ and N₂O emissions from paddy plots generally fall within the range of emissions in other studies (Jiang et al. 2009; Yao et al. 2012). However, the emissions were higher

Fig. 4 Seasonal variation of nitrous oxide (N₂O) fluxes from orchard (a) and paddy (b) soils. The data are shown as means with standard errors. The black arrow indicates the cultivation of orchard and paddy. The light gray arrows in panel a indicate fertilizer applied to OF, and the gray arrows in panel b indicate fertilizer applied to PF

Fig. 5 Cumulative CH₄ (a, b) and N₂O (c, d) fluxes measured from the four treatments after conversion. The data are shown as means

than those reported in the subtropical paddy fields nearby the study site (Zhang et al. 2010) and other paddy fields in China (Xie et al. 2010; Yao et al. 2012; Tang et al. 2015). Thus, it can be presumed that the discrepancy in CH₄ and N₂O fluxes from paddy soils may be the disparate fertilizer rate, water regime, and cropping systems. Besides, the above former studies mainly focused on the CH₄ and N₂O emissions from the growing season; therefore, an alternative explanation for the higher emissions could be the neglect of the emissions from the fallow season.

The soil CH₄ uptakes and N₂O fluxes of newly converted orchard corresponded well with previous results (Iqbal et al. 2009; Rowlings et al. 2013; Zona et al. 2013). However, annual CH₄ uptakes of newly converted orchard were significantly lower than those observed by Liu et al. (2008) who reported an average annual soil CH₄ uptake of 2.61 kg C ha⁻¹ year⁻¹ in the subtropical region of southern China.

This difference can be attributed to the discrepancy in the orchard establishment years (12 years in south China versus 1 year in red soil). As it was verified that orchards had been established for many years in subtropical, China consumed atmospheric CH₄ at much higher rates because of less soil disturbance (Iqbal et al. 2009). Similarly, in a temperate spruce forest, Wu et al. (2011) found that the reduced soil CH₄ oxidation capacity due to land use conversion could last for a decade or even longer. Thus, long-term continuous measurements over several years are needed to understand the dynamics of GHG fluxes during different stages after land use conversion. Conversely, annual N₂O emissions (15.94 and 9.48 kg N ha⁻¹ year⁻¹) from orchard were significantly higher than those in humid subtropical Australia (Rowlings et al. 2013), mid-subtropical China (Lin et al. 2012), and south China (Liu et al. 2008), although the amounts of applied N fertilizer rate (176 kg N ha⁻¹ year⁻¹) fall in the range of the

Table 4 Effects of land use conversion and fertilization on soil cumulative CH₄, N₂O emissions, and CO₂ equivalent 1 year after conversion

Gas	Cumulative emissions				Relative change (%)			
	OF	ONF	PF	PNF	OF PF -1	ONF PF -1	PF PNF -1	OF ONF -1
CH ₄ (kg C ha ⁻¹ year ⁻¹)	-0.76 ± 0.17b	-0.73 ± 0.22b	355.86 ± 179.62a	406.25 ± 114.16a	-100	-100	-12	4
N ₂ O (kg N ha ⁻¹ year ⁻¹)	15.94 ± 0.72a	9.48 ± 0.64b	3.06 ± 0.32c	1.43 ± 0.19d	421	210	114	68
CO ₂ eq (kg C ha ⁻¹ year ⁻¹)	2026.73 ± 91.1b	1201.61 ± 83.2c	4790.53 ± 339.2a	5205.36 ± 172.9a	-58	-75	-7	69

The data are shown as means with standard errors. Different letters in the same row indicate significant differences ($P < 0.05$) between corresponding treatments

Table 5 Pearson correlation coefficients between soil CH₄ and N₂O fluxes with soil properties in the orchard and paddy plots

Land use	Fluxes	Soil temp. (°C)	Soil water content (%)	NH ₄ ⁺ -N (mg kg ⁻¹)	NO ₃ ⁻ -N (mg kg ⁻¹)	DOC (mg l ⁻¹)
Orchard	CH ₄ (μg C m ⁻² h ⁻¹)	0.23	0.69**	0.21	-0.24	0.88**
	N ₂ O (μg N m ⁻² h ⁻¹)	0.35**	-0.31	0.22	0.86**	-0.246
Paddy	CH ₄ (μg C m ⁻² h ⁻¹)	0.44**	—	0.54**	-0.14	0.86**
	N ₂ O (μg N m ⁻² h ⁻¹)	0.19	—	-0.06	0.86**	-0.04

*Significant correlation at the level of 0.05, **significant correlation at the level of 0.001

above studies (120–210 kg N ha⁻¹ year⁻¹). The higher N₂O emissions from newly orchard plots were probably related to higher amounts of basal fertilizer (370 kg N ha⁻¹ year⁻¹). Thus, these results suggested that continuous measurements of soil CH₄ and N₂O fluxes in the early days after conversion are needed to reliably estimate emissions from converted orchard, which will be helpful for the global budget of GHG emissions induced by land use conversion.

Effects of land use conversion on soil CH₄ and N₂O fluxes

In our study, land use conversion from paddy to orchard reduced CH₄ emissions and increased N₂O emissions by 100 and 421 %, respectively (Table 3). The dynamics of carbon and nitrogen in converted orchard fields significantly differed from those in paddy fields in which anaerobic decomposition process was dominant. It has been reported that CH₄ was actively produced in the anaerobic soil conditions with the preference of methanogens and emitted to the atmosphere through the rice plants (Bhattacharyya et al. 2013; Nazaries et al. 2013). Conversely, anaerobic conditions in the paddy plots limited nitrate availability, and anaerobiosis favored denitrification to N₂ (Zou et al. 2007), thus leading to the lower N₂O emissions. However, in newly converted upland orchard, the regeneration of oxidants, in particular re-oxidation of Fe(II), thus contributes to the uptake of CH₄ (Eusufzai et al. 2010). Meanwhile, N₂O emissions were generally higher in optimum soil moisture and N supply as a result of tight coupling between nitrification and denitrification (Snyder et al. 2009). Those results were in well agreement with earlier studies (Knox et al. 2015; Weller et al. 2016) showing that the formation of aerobic conditions caused by land use conversion could result in the reduced CH₄ fluxes at the expense of increasing N₂O fluxes (Jiang et al. 2009). In our study, episodic higher CH₄ emission still occurred in the early conversion

days (Fig. 3a), possibly owing to a relocation of the methanogens further down the soil profiles. According to the former literature and results obtained in our study, we argued that soil conditions induced by land use conversion might be the main controlling factors influencing soil CH₄ and N₂O emissions, which should be highly verified in subsequent studies.

Meanwhile, higher soil labile organic substrates (especially DOC) were generally found to increase CH₄ production because they could provide substrates for methanogens (Zou et al. 2005; Hu et al. 2016). In the present study, based on the agricultural practices, some crop residue was typically retained in paddy fields, which led to high level of soil DOC concentration throughout the observing period (Table 2). While in orchard fields, soil DOC contents decreased due to the improved soil aeration and increased mineralization activity (Weller et al. 2016). Thus, our result confirmed that higher CH₄ fluxes were observed from paddy soils where DOC contents were higher (Zou et al. 2005; Yao et al. 2012). Studies also showed that the higher level of DOC contents may have enhanced C availability thus controlled N₂O emissions through its influence on nitrify and denitrifying activity (Burger et al. 2005). Here in our study, N₂O emissions from orchard plots were higher than those in the paddy plots, while reverse pattern for DOC concentrations. The contrasting findings, however, point out that the DOC concentrations cannot be the key determinant of the soil N₂O fluxes induced by land use conversion.

The fluxes of CH₄ and N₂O are influenced by soil properties, such as soil temperature and pH (Iqbal et al. 2009; Lin et al. 2012). The conversion of paddy to orchard caused the soil temperature to be higher in the orchard than that in the paddy plots (Table 2), which was reported to accelerate both nitrification and denitrification process (Saggar et al. 2008) and thus led to more N₂O emissions from orchard plots (Table 4).

Table 6 Multiple regression analysis (stepwise) between soil CH₄ and N₂O fluxes and soil properties in the four treatments

Land use	Fluxes	Regression model	R ²	P value
Orchard	CH ₄ (μg C m ⁻² h ⁻¹)	$Y = 147.24 \text{ DOC} - 3530.18$	0.77	<0.001
	N ₂ O (μg N m ⁻² h ⁻¹)	$Y = 31.67 \text{ NO}_3^- \text{-N} - 4.391$	0.73	<0.001
Paddy	CH ₄ (μg C m ⁻² h ⁻¹)	$Y = 143.06 \text{ DOC} - 4491.39$	0.74	<0.001
	N ₂ O (μg N m ⁻² h ⁻¹)	$Y = 90.90 \text{ NO}_3^- \text{-N} - 55.79$	0.74	<0.001

Besides, soil pH is also an important factor regulating N_2O emissions, as former studies have reported that higher soil pH could potentially favor the activity of N_2O reductase from denitrifying microorganisms (Wang et al. 1993). In our study, soil pH of paddy fields was significantly higher than that of orchard (data are not shown) and promotes the process from N_2O to N_2 , thus decreasing N_2O emissions from paddy soils. In summary, the difference in soil properties could be responsible for the fact that the CH_4 and N_2O fluxes differed after conversion.

Effects of fertilization on soil CH_4 and N_2O fluxes

Generally, the effects of N fertilization on soil CH_4 flux are controversial, including positive (Krüger and Frenzel 2003), negative (Zhou et al. 2016), and neutral effects (Fan et al. 2016) in different land uses. In this study, we found that fertilization did not significantly change soil CH_4 uptakes in orchard plots but could decrease CH_4 fluxes in paddy plots (Table 4), though soil $\text{NH}_4^+\text{-N}$ concentrations significantly increased after fertilizer application (Fig. 2a). According to the former statement (Schimel 2000), the inhibition or stimulation mechanisms induced by fertilization in the upland fields were different from those in paddy plots. Generally, N fertilizers applied to upland soils soon hydrolyzes into $\text{NH}_4^+\text{-N}$, which led to compete for the same active sites of CH_4 monooxygenases and thus inhibited CH_4 oxidation (Wang et al. 2015). While in paddy fields, fertilization could stimulate CH_4 production and vascular transport capacity via enhancing methanogenic growth/activity and rice growth (Schimel 2000). Moreover, fertilization was advantageous to the methanotrophs and some other methane oxidation bacteria growth and then accelerated methane oxidation in both upland fields and paddy fields (Schimel 2000; Reay and Nedwell 2004). Thus, an alternative explanation for the neutral effects of fertilization on soil CH_4 fluxes from orchard plots could be the potential inhibition of CH_4 uptakes from nitrogen addition being compensated by nitrogen stimulation on the substrates. In addition, these results also implied that the selective fertilizer ($356 \text{ kg N ha}^{-1} \text{ year}^{-1}$) in paddy fields, which resulted in a markedly larger stimulation of CH_4 oxidation than the CH_4 production, would be beneficial for mitigation of soil CH_4 emissions.

The N_2O emissions from soil are mainly controlled by the process of nitrifying and denitrifying, which can be strongly affected by fertilization (Yao et al. 2012; Rowlings et al. 2013). The higher mineral N contents, which was increased by fertilization, provide more substrates for nitrification and denitrification and thus stimulated soil N_2O fluxes. Meanwhile, previous studies also stated that soil DOC contents could control soil N_2O emissions by influencing microbial growth and biological decomposition (Zou et al. 2005). In our present study, higher soil DOC contents and mineral N

contents together predicated more N_2O emissions from fertilized treatment compared to the unfertilized treatment (Table 4).

Dependence of soil CH_4 and N_2O on soil properties

As expected, soil CH_4 flux was distinctly affected by soil properties (Table 5). Soil temperature is widely reported to be positively correlated with CH_4 fluxes in orchard (Liu et al. 2008) and upland crops (Iqbal et al. 2009). In our study, such significant positive relationships existed in paddy plots, suggesting that soil temperature was one of the main driving forces of soil CH_4 emissions. However, no significant relationships between them were observed in orchard plots. Hence, the temperature, at which methanotroph activity was stable, differed across land use types and water conditions (Trotsenko and Khmelenina 2005). Meanwhile, soil water content was usually considered to be another important regulator of CH_4 fluxes or uptakes (Zou et al. 2005; Wang et al. 2015). Sustainable water logging would favor the existence of methanogens and led to more CH_4 fluxes (Zou et al. 2005), which is also supported by our results (Figs. 1b and 3b). Significant positive relationships between CH_4 fluxes and soil water content were also observed in orchard plots (Table 5). In general, the optimal soil moisture for methane oxidation is approximately 10–20 % water-filled pore space (WFPS) (Lin et al. 2012). Low soil moisture would inhibit the activity of methanotrophs, and there is also a negative effect on methane oxidation when the soil moisture is too high because water blocks the diffusion channel of methane and oxygen in the soil (Lin et al. 2012). Moreover, increased amount of inorganic N pools ($\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$) has been showed to reduce or increase soil CH_4 fluxes (Iqbal et al. 2009; Wang et al. 2014). In our study, CH_4 fluxes had a significant positive relationship with soil $\text{NH}_4^+\text{-N}$ in paddy plots, which was not in agreement with the Bodelier et al. (2000), who found that the fertilizer addition decreased CH_4 fluxes. Probably, the controversial results might be related to the fertilizer rate. Besides, a strong dependence of soil CH_4 on soil DOC concentration in paddy and orchard fields was observed, which was similar to results obtained in other regions (Zou et al. 2005; Hou et al. 2013). Overall, multiple regression models indicated the soil DOC contents were the main regulating parameter of the CH_4 fluxes in both orchard and paddy plots.

N_2O fluxes displayed pronounced dependence on soil properties from the two land use types (Table 5). Soil temperature exhibited distinctive positive correlation with N_2O fluxes in orchard fields (Table 5), which was in well agreement with the results from other studies (Liu et al. 2008; Lin et al. 2010). For paddy soils, soil temperature was not a sensitive factor to regulate N_2O fluxes during our observing period. Generally, temperature regulated the N_2O emissions through its influence on nitrifying and denitrifying activity

(Burger et al. 2005). However, this was in contrast with the finding from a similar study by Tang et al. (2006), who only reported a reasonable correlation between soil moisture and N_2O emissions. In our study, statistical positive relationship between N_2O emissions and soil moisture were detected in orchard plots, which were in line with the former studies (Tang et al. 2006; Liu et al. 2008). Nevertheless, the stimulation of soil moisture on N_2O fluxes may be significant at dry soils and wet soils (Lin et al. 2012). Besides, several studies have implied that the amount of DOC is a measure of the readily available resource for microbial growth and biological decomposition (Zou et al. 2005; Lin et al. 2012). On the other hand, the N_2O fluxes from paddy and orchard plots could not be correlated to DOC contents (Table 5). Lin et al. (2012) also found that the relationship between DOC contents and soil N_2O fluxes had strong dependence on soil moisture, showing that the DOC contents explained up to 49 % of soil N_2O flux when WFPS was higher than 45 %, while it only accounted for 6 % of N_2O flux when WFPS was lower than 45 %. Generally, soil inorganic N availability (especially NO_3^- -N) was the most important regulating factor for magnitude of emissions under the orchard and paddy, which has been widely discussed before (Lin et al. 2010; Yao et al. 2012; Rowlings et al. 2013). Therefore, for precise estimation of N_2O emissions from different land use, quantitative evaluation of NO_3^- -N is necessary, which serves as the substrates for denitrifying.

Combined climate effects of CH_4 and N_2O emissions

As mentioned above, even if the CH_4 emissions decreased coupling with land use conversion, the increases in N_2O emissions would produce a trade-off in terms of the greenhouse effect. Recently, the net GWPs have been estimated to complete understanding of land use conversion's impact on radiative forcing. In the preset study, land use conversion from paddy to orchard significantly decreased the net GWPs irrespective of fertilization (Table 4). These results suggest that orchard cultivation could significantly mitigate the integrative greenhouse effect caused by CH_4 and N_2O compared with paddy fields, which was in well agreement with the results obtained from other studies (Scheer et al. 2008; Kong et al. 2013). The higher GWPs from paddy fields were mainly related to the CH_4 fluxes in the flooding period. The observed GWP estimates for paddy fields were much higher than those observed in the Khorezm region, Germany, that summarized that the seasonal GWPs of the different cropping sites ranged from 200 to 3000 $\text{kg C ha}^{-1} \text{ year}^{-1}$ (Scheer et al. 2008). Compared to the ONF, the higher GWPs were observed in the OF, which was explained by the high input of mineral N and resulting in high N_2O emissions (Scheer et al. 2008). Fertilizer applied to the paddy fields could decrease GWPs, but the values were not statistically significant, which is consistent with the theory of Zou et al. (2005) who summarized

that the fertilizer application generally depressed or did not affect the net GWPs depending on the fertilizer rate. It is noteworthy that the overall GHG balance estimates should include the net exchanges of CO_2 , N_2O , and CH_4 between agroecosystems and the atmosphere. However, the net exchange of CO_2 between agroecosystems and atmosphere was not examined in this study. With consideration of results in the literature, rice paddy was usually found to be a weak atmospheric CO_2 sink (e.g., Knox et al. 2015), and this carbon sink strength was normally smaller than that of mature orchard (Zanotelli et al. 2015). However, Knox et al. (2015) found that the drained agriculture site were net carbon and GHG sources, indicating that there are still many uncertainties in net exchange of CO_2 between agroecosystems and atmosphere during land use conversion. Thus, further research should include measurements of the climatically important C and N trace gas fluxes (CO_2 , N_2O , and CH_4) to provide a complete evaluation of the overall GHG balance during land use conversion.

Conclusions

To our knowledge, the reported datasets represented the first detailed CH_4 and N_2O fluxes from red soils during different stages after land use conversion from paddy to orchard in mid-subtropical China. The results showed that current shift in agricultural land use from conventional paddy fields to upland orchards significantly decreased soil CH_4 fluxes but increased soil N_2O fluxes. As a result, we demonstrated that the GWPs would decrease if converting paddy to orchard. Soil temperature, soil water content, soil DOC, NH_4^+ -N, and NO_3^- -N concentrations also varied and were strongly related to variation of soil CH_4 and N_2O fluxes. In addition, fertilization significantly decreased the CH_4 fluxes from paddy soils but failed to affect the CH_4 fluxes from orchard soils, whereas fertilization distinctly increased the N_2O fluxes from both orchard soils and paddy soils. Thus, fertilization increased GWPs of orchard soils but did not significantly affect GWPs of paddy soils. To be concluded, these findings underscore the importance of land use conversion and selective fertilization concerning inventories and mitigation options for GHGs. Moreover, to accurately estimate national and global CH_4 and N_2O emissions induced by land use conversion, direct field measurements in the early stages after conversion are highly needed.

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