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The contrasting effects of deposited NH_4^+ and NO_3^- on soil CO_2, CH_4 and N_2O fluxes in a subtropical plantation, southern China

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ABSTRACT

Background and aims: Deposited NH_4^+ and NO_3^- differently affect soil carbon (C) and nitrogen (N) cycles due to their contrasting actions in terrestrial ecosystems. However, little information on the effects of exogenous NH_4^+ and NO_3^- inputs on the exchange of greenhouse gases (GHGs) from the subtropical plantation soils as well as their contribution to global warming is available to date.
Methods: Based on a field experiment, two-form (NH_4Cl and $NaNO_3$) and two-level (40 and 120 kg N ha^{-1} yr^{-1}) of N addition, in a slash pine plantation of southern China, we investigated soil CO_2, CH_4 and N_2O fluxes and related auxiliary variables (soil temperature and moisture) twice a week using static chamber–gas chromatography. The total global warming potential (GWP) of soil GHG fluxes and N_2O emission factor (EF) were calculated.
Results: Low level of $NaNO_3$ addition significantly increased cumulative annual soil CO_2 emission by 33.7%. N addition significantly promoted annual soil N_2O emission by 2.4–6.9 folds; moreover, ammonium-N addition had a greater promotion to soil N_2O emission than nitrate-N addition. However, short-term N addition did not change soil CH_4 uptake. Also, soil CO_2 and N_2O fluxes were positively correlated with soil temperature and moisture, while soil CH_4 uptake was only driven by soil moisture. Overall, elevated N addition increased the total GWP, and changed the temperature sensitivity (Q_{10}) of soil CO_2 and N_2O fluxes.
Conclusions: These results suggest that chronic atmospheric N deposition changes soil-atmospheric GHG fluxes in the subtropical plantation of southern China depending on the levels and forms of N input, and would exacerbate global warming.

1. Introduction

Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are greenhouse gases (GHGs) that regulate global warming. Currently, the concentrations of CO_2, CH_4 and N_2O in the atmosphere are increasing by 0.5% yr^{-1}, 0.9% yr^{-1}, and 0.3% yr^{-1}, respectively, and they contribute to more than 87% of global warming (Foster et al., 2007). Terrestrial ecosystems are vital regulators of atmospheric GHG concentrations with a considerable amount of them produced and consumed through soil processes (Canadell et al., 2007). The exchanges of CO_2, CH_4 and N_2O from global land ecosystems are estimated at -2.8 Pg C yr^{-1}, 144.39 Tg C yr^{-1} and 12.52 Tg N yr^{-1}, respectively (Battin et al., 2009; Tian et al., 2013). However, the exchanges of CO_2, CH_4, and N_2O between soil and atmosphere are highly variables, depending on a number of environmental factors and soil properties (Dalal and Allen, 2008). Climate change, as an anthropogenic disturbance, can also impact and feedback soil GHG fluxes (Dijkstra et al., 2013). To accurately predict the changes of our future environment, it is necessary to better understand the cause of these variations.

The production and consumption of CO_2, CH_4 and N_2O in soils are microbe-mediated processes and closely coupled under natural conditions (Thornton et al., 2009). Typically, CO_2 is mainly produced by the decomposition of organic matter and the process of root respiration. The methanotrophs in aerobic soil can oxidize atmospheric CH_4 into CO_2, while N_2O is a byproduct of nitrification ($NH_4^+ \rightarrow NO_3^-$) and denitrification ($NO_3^- \rightarrow N_2$). Globally,

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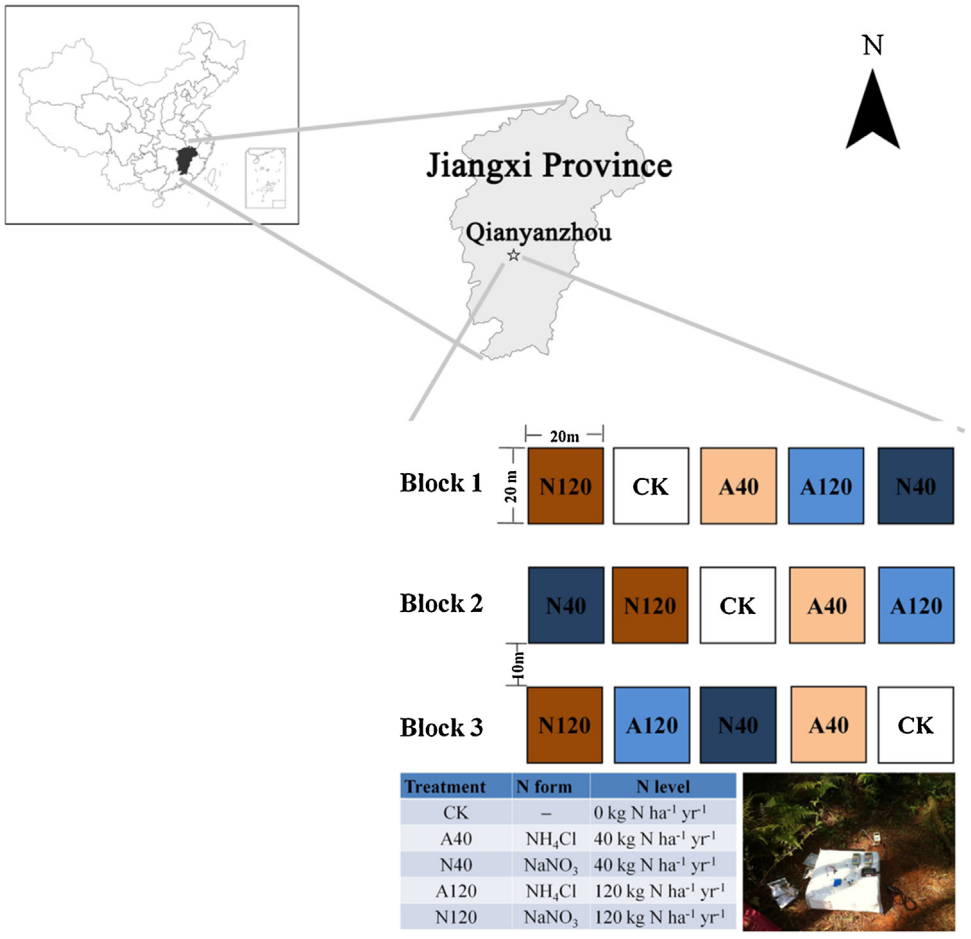
anthropogenic atmospheric N deposition into the biosphere has increased by more than three-fold since 1860s (Galloway et al., 2008), thereby promotes vegetation production (Magnani et al., 2007), decreases soil CO_2 emission and CH_4 uptake (Zhang et al., 2008b), and increases soil N_2O emission (Lu et al., 2011), although some contrary effects and lacks of response have also been reported (Ambus and Robertson, 2006; Fang et al., 2012; Xu et al., 2014). Considering the effects of N deposition on soil CH_4 uptake and N_2O emission, the carbon (C) sequestration potential elicited by N deposition would be offset by more than 50% (Liu and Greaver, 2009). Unfortunately, the above understanding mainly originates from the undisturbed temperate forests in the North America and Europe, little information is available regarding the responses of soil CO_2, CH_4 and N_2O fluxes in the subtropical forests to increased N deposition (Yan et al., 2014). Also, most studies have been conducted using single N fertilizer type (e.g., NH_4NO_3 or urea) (Mo et al., 2008; Phillips and Podrebarac, 2009), and the contrasting effects of oxidized NO_3^- and reduced NH_4^+ have rarely been investigated (Wang et al., 2014).

Forest plantations cover approximately 200 million ha in globe (FAO, 2007). Their total C storage is 11.8 Pg C and increases at a rate of 0.18 Pg C yr^{-1}, playing very important roles in the sequestration of atmospheric CO_2 (Winjum and Schroeder, 1997). China has the largest plantation area (62 million ha) in the world (Piao et al., 2009; Chen et al., 2011), approximately 63% of plantations in China distribute in the subtropical region where N deposition rate is the highest (>30 kg N ha^{-1} yr^{-1}) (Zhu et al., 2015). The subtropical plantation forests are vulnerable to increased N deposition due to single community structure and barren soil fertility. Previous studies show that simulated NH_4NO_3 inputs to the subtropical forests significantly inhibit litter decomposition and soil CO_2 emission (Fang et al., 2007; Mo et al., 2008), decrease CH_4 uptake (Zhang et al., 2008b, 2012b), and increase gaseous N emission (N_2O, NO, and N_2) (Zhang et al., 2008a, 2009). Also, deposited NH_4^+ is immobilized by soils and is assimilated by plants and microbial communities, whereas plants use NO_3^- more than NH_4^+ due to its high mobility (Sheng et al., 2014). The contrasting effects of N cumulation on root autotropical respiration and microbial heterotropical respiration dominate the reponses of soil CO_2 flux to N addtion (Wang et al., 2015). Furthermore, soil NH_4^+ cumulation generally reduces CH_4 oxidation (Bodelier, 2011), and the effects of soil NO_3^- on soil CH_4 uptake are controverial incluing promotion and inhibition (Wang et al., 2014; Xu et al., 2014). Exogenous N inputs regulate forest soil CH_4 uptake through the competitive inhibition and toxic inhibition, and increase soil N_2O emission through promoting soil nitrification and denitrification processes (Fang et al., 2014). Therefore, we hypothesized that (1) increased NO_3^- deposition would promote soil CO_2 emission through increasing root heterotrophic respiration, and (2) NH_4^+ deposition would decrease soil CH_4 uptake and increase N_2O emission, more so than NO_3^- deposition.

To test the two hypotheses, we conducted a two-form and two-level N addition experiment in a coniferous forest plantation of subtropical China. The specific aims of the study were (1) to examine the effects of simulated NH_4^+ and NO_3^- deposition on soil GHG fluxes and total global warming potential (GWP) and (2) to evaluate the relationships between soil GHG fluxes and hydrothermal factors and between cumulative GHG fluxes and N addition levels.



Treatment	N form	N level
CK	–	0 kg N ha^{-1} yr^{-1}
A40	NH_4Cl	40 kg N ha^{-1} yr^{-1}
N40	$NaNO_3$	40 kg N ha^{-1} yr^{-1}
A120	NH_4Cl	120 kg N ha^{-1} yr^{-1}
N120	$NaNO_3$	120 kg N ha^{-1} yr^{-1}

Fig. 1. Location of study site and illustration of field plots in the N addition experiment.

2. Materials and methods

2.1. Study site

This study was conducted at the Qianyanzhou Ecological Station of Chinese Academy of Sciences (26°44′39″N, 115°03′33″E), situated on the typical red earth hilly region in Taihe city, Jiangxi province in mid-subtropical China (Fig. 1). The topography is missing from gently undulating with an average slope of 10–30°. The warm and humid monsoon climate dominates the region. The highest and lowest extreme temperatures are 39.5 °C and −5.8 °C, respectively, with a mean annual temperature of 17.9 °C. The mean annual precipitation amounts to about 1505 mm, and 51% and 30% occurred between March and June and between July and October, respectively. The vegetation is dominated by coniferous forest plantation, planted around 1985. The main canopy species are splash pine (*Pinus massoniana*), masson pine (*Pinus elliottii*), and cedarwood (*Cunninghamia lanceolata*). Soils are typical red soils, classified as Cambosols, which are derived from sandstone and sandy conglomerate (IUSS Working Group, 2006). Topsoil (0–20 cm) organic C is 20.44 g kg^{-1}, total N is 1.10 g kg^{-1}, total phosphorus is 1.12 g kg^{-1}, pH is 4.26, and soil bulk density is 1.54 g cm^{-3} (Wang et al., 2014).

2.2. Experimental design

The subtropical slash pine plantation was selected to perform N addition experiment. The measured rate of atmospheric N deposition is 32.6 kg N ha^{-1} yr^{-1} in 2013, and the NH_4^+/NO_3^- ratio is 6/5 (Zhu et al., 2015). The experiment was a random block design with three replicates. To simulate the effects of deposited NH_4^+ and NO_3^- on soil C and N biogeochemical cycles and ecosystem C sequestration, two kinds of fertilizers (NH_4Cl and $NaNO_3$) were added at two rates: 40 and 120 kg N ha^{-1} yr^{-1}. A control was set at each block. Five treatments were hereafter referred to as CK, A40, N40, A120, and N120, respectively (Fig. 1). A total of 15 plots (20 m × 20 m) were established, each surrounded by 10 m-wide buffer strip. N fertilizer solutions were sprayed below the canopy on a sunny day or after the rain during the first week of every month, and the control plots only received equivalent amounts of water (Wang et al., 2014). The N addition experiment was conducted over 3 years beginning in May 2012 and until now.

2.3. Measurements of soil GHG fluxes and auxiliary variables

Soil CO_2, CH_4 and N_2O fluxes were collected in opaque static chambers and measured using gas chromatography. The static chambers are assembly consist of a permanently installed stainless steel base (length × width × height = 50 cm × 50 cm × 10 cm) and a removable top (length × width × height = 50 cm × 50 cm × 15 cm). The available volume of chamber was 37.5 L. Five gas samples were taken from the chamber head-space over 40 min between 9:00 and 11:00 am (China Standard Time, CST), and then analyzed for GHG concentrations on a gas chromatograph (Agilent 7890A, USA) within 24 h. Soil GHG fluxes were calculated from the slope of linear or nonlinear regression between each GHG's concentration and time (Zheng et al., 2008). The correlation coefficient of the regression had to be significant at a 95% confidence limit for $n = 5$ ($r^2 = 0.95$), otherwise the sample was rejected. Soil GHG fluxes were measured in each plot eight times a month, and dense gas collection (4 times) was performed in the first week of every month to capture the peak of soil N_2O emission (Wang et al., 2014).

Meanwhile, soil temperature (Ts) and soil moisture (Ms) measurements were performed at a soil depth of 10 cm, at random locations within each plot, using a portable temperature probes (JM624 digital thermometer, Living–Jinming Ltd., China) and a moisture probe meter (TDR100, Spectrum, USA).

2.4. Calculation of soil GHG fluxes data

The annual amount of soil GHG fluxes can be calculated using the Eq. (1) (Jiang et al., 2010):

$$E_{GHG} = \frac{\sum_{i=1}^{n} 0.5 \times (F_i + F_{i+1}) \times (t_{i+1} - t_i) \times 24}{100000} \quad (1)$$

where, E_{GHG} is the cumulative soil GHG flux (t CO_2–C ha^{-1} yr^{-1}, kg CH_4–C ha^{-1} yr^{-1}, or kg N_2O–N ha^{-1} yr^{-1}, respectively), F is the net change flux of soil-atmospheric CO_2, CH_4, and N_2O (mg CO_2–C m^{-2} h^{-1}, μg CH_4–C m^{-2} h^{-1}, and μg N_2O–N m^{-2} h^{-1}) determined at each sampling time, i is the sampling number, and t is the sampling time based on the Julian day.

Global warming potential (GWP) is an index defined as the cumulative radiative forcing between the present and some chosen later time scale caused by a unit mass of gas emitted during the present condition (Robertson et al., 2000). It is used to compare the effectiveness of each GHG to trap heat in the atmosphere relative to some standard gas, by convention CO_2. The total GWP of GHG exchanges (t CO_2-eq. ha^{-1} yr^{-1}) is obtained using Eq. (2):

$$GWP = F_{CO_2} + \gamma_{CH_4} \cdot F_{CH_4} + \gamma_{N_2O} \cdot F_{N_2O} \quad (2)$$

where, F_{CO_2}, F_{CH_4}, and F_{N_2O} are the net exchange rates (t species ha^{-1} yr^{-1}) of CO_2, CH_4, and N_2O, respectively; γ_{CH_4} and γ_{N_2O} are the radiative forcing effects of CH_4 and N_2O relative to that of CO_2, and are equal to 25 and 298 over a 100-year period, respectively (Foster et al., 2007).

Emission factor (EF, %) links between the amount of N fertilizer applied and N_2O emission, and is calculated using the Eq. (3).

$$EF = \frac{AE_i - AE_0}{N\ \ Rate_i} \times 100\% \quad (3)$$

where, AEi is the cumulative N_2O emissions from i treatment (kg N_2O–N ha^{-1}), AE_0 is the cumulative N_2O emissions from control treatment (kg N_2O–N ha^{-1}), and $N\ Rate_i$ is the N fertilization rate in i treatment (kg N ha^{-1}).

The sensitivity of soil CO_2 flux to soil temperature at 10 cm depth (Q_{10}) is obtained from a coefficient, k, in the exponential equation (Eqs. (4) and (5)) (Lloyd and Taylor, 1994):

$$F_{CO_2} = a \times e^{kt} \quad (4)$$

$$Q_{10} = \frac{a \times e^{k(10+t)}}{a \times e^{kt}} = e^{10k} \quad (5)$$

where, F_{CO_2} is the soil CO_2 flux, t is the soil temperature, and k is a regression coefficient.

2.5. Statistical analysis

We used a repeated measures analysis of variance (ANOVA) to evaluate the differences in soil GHG fluxes and auxiliary variables among the different forms and levels of N addition. Comparisons of the means were conducted using the Tukey's Honestly Significant Difference (HSD) test. We used one-way ANOVA with HSD test to evaluate the effects of experimental treatments on cumulative annual GHG amount, GWP value, and EF value. Standardized regression analysis was used to examine the relationships between soil GHG fluxes and environmental factors with the N addition levels as categorical variables (Fang et al., 2012). All statistical analyses were performed using the SPSS software package (version 16.0). Statistical significant differences were accepted at $p < 0.05$ unless otherwise stated.

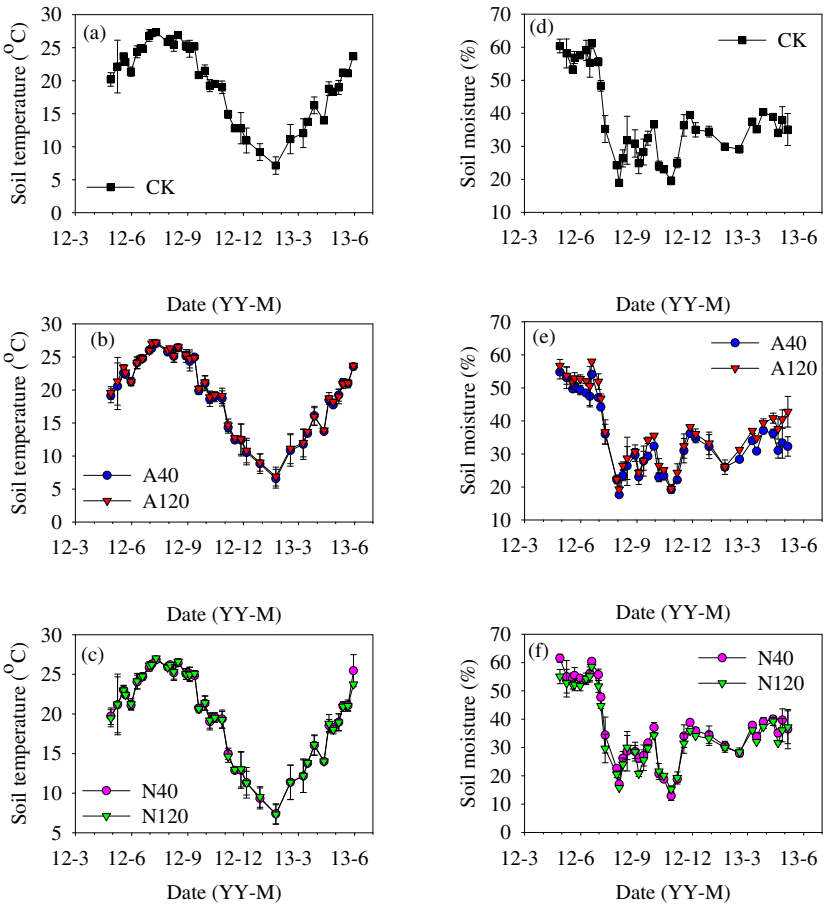


Fig. 2. Monthly variation of soil temperature and soil moisture at 0–10 cm depth under different experimental treatments. Data are shown as means with standard errors ($n = 6$).

3. Results

3.1. Soil temperature and moisture

There were significant seasonal variations in soil temperature (Ts) and moisture (Ms) at 0–10 cm depth under the five treatments (Fig. 2, Table 1, $p < 0.001$). The maximum (27.3 °C) and minimum (7.2 °C) of 10 cm Ts occurred at the mid-July and at the end of January, respectively (Fig. 2(a)–(c)). Soil moisture at 0–10 cm depth ranged from 18.95% to 61.14% with the maximum occurring in the plum rain season of May (Fig. 2(d)–(f)). Significant difference in Ts and Ms among the three blocks was observed, too (Table 1, $p = 0.03$ and $p < 0.001$ for Ts and Ms, respectively). Also, neither the levels nor the forms of N addition affected Ts, but they tended to decrease Ms for a certain observation (Table 1). A40 and N120 treatments significantly decreased Ms in comparison with control (Table 2).

3.2. Soil GHG fluxes

Similar to Ts, soil CO_2 flux exhibited a single-peak and single-trough pattern, with the minimum and maximum occurring in mid-June and early January, respectively ($p < 0.001$, Fig. 3(a)–(c)). Soil CO_2 flux fluctuated from 9.18 to 120.89 mg C m^{-2} h^{-1} under the control, with an average of 59.53 ± 0.81 mg C m^{-2} h^{-1} (Table 2). This rate was translated into a cumulative annual soil CO_2 emission of 4.66 ± 0.97 t C ha^{-1} yr^{-1} (Table 2). N level rather than N form significantly changed soil CO_2 flux, and their interaction was significant, too (Table 1, $p < 0.001$). N40 treatment significantly increased the average soil CO_2 flux and its annual cumulative emission by 36.87% and 47.21%, respectively (Table 2).

Soil CH_4 flux showed an obvious seasonal volatility with CH_4 uptake and emission alternatively occurring ($p < 0.001$, Fig. 3(d)–(f)). Overall, the subtropical plantation soil was a sink for atmospheric CH_4. The average soil CH_4 fluxes at the five treatments ranged from −6.13 to −3.23 μg C m^{-2} h^{-1} (Table 2). N level did not significantly affect soil CH_4 flux (Table 1, $p = 0.84$). However, N form slightly changed soil CH_4 flux at the level of 0.1 (Table 1, $p = 0.10$), and the difference between N40 and A120 treatments was significant (Table 2). One-year N addition did not significantly change the annual cumulative CH_4 uptake in the subtropical plantation soils (Table 2).

The seasonality of soil N_2O flux was significant with the maximum and minimum occurring in summer and in winter, respectively (Fig. 3(g)–(i), Table 1, $p < 0.001$). Both N level and N form significantly influenced soil N_2O fluxes (Table 1, $p < 0.001$). The average soil N_2O flux at the four N addition treatments ranged from 42 to 96.78 μg N m^{-2} h^{-1}, which was 4–11 times higher than that of control (Table 2). A40, N40, A120, and N120 treatments significantly increased the annual cumulative N_2O emission by 364%, 236%, 687%, and 519%, respectively relative to the control; moreover, the increasing percentage was higher from ammonium-N fertilizer than from nitrate-N fertilizer at the same level of N addition (Table 2). In addition, the average EF values decreased with the N level ranging from 4.10 to 8.67, and ammonium-N fertilizer had a greater EF value than nitrate-N fertilizer (Table 2).

Regardless of the experimental treatments, the CO_2 was the most influential GHG in increasing the total GWP. The contribution

Table 1
Results of repeated measures ANOVA on the effects of date, N level, N form, and their interactions on soil CO_2, CH_4 and N_2O fluxes and related soil variables.

Source of variation	Soil temperature		Soil moisture		Soil CO_2 flux		Soil CH_4 flux		Soil N_2O flux	
	F	*P*	*F*	*P*	*F*	*P*	*F*	*P*	*F*	*P*
Between subjects										
Block	3.79	0.03	10.37	0.001	8.22	<0.001	1.04	0.36	12.91	<0.001
N level	1.59	0.21	1.04	0.37	15.14	<0.001	0.18	0.84	35.79	<0.001
N form	1.99	0.17	1.38	0.25	0.84	0.37	2.81	0.10	16.40	<0.001
N level × N form	0.53	0.47	3.08	0.09	31.57	<0.001	<0.001	0.98	0.06	0.80
Within subjects (multivariate)										
Date	418.90	<0.001	40.41	<0.001	120.88	<0.001	4.15	<0.001	34.04	<0.001
Date × Block	4.34	<0.001	4.01	<0.001	2.09	0.01	0.88	0.62	2.03	0.01
Date × N level	1.54	0.08	2.36	0.02	2.00	0.02	0.66	0.86	4.48	<0.001
Date × N form	1.98	0.05	4.06	0.01	1.34	0.26	1.77	0.09	2.69	0.01
Date × N level × N form	0.645	0.78	1.70	0.19	2.27	0.04	0.91	0.54	1.11	0.38

of CO_2 flux to the total GWP was more than 70%, whereas the contributions of CH_4 and N_2O fluxes were less than 30% (Table 2). N addition consistently increased the total GWP value, and N40 and A40 treatments had the greatest promotion (Table 2).

3.3. Relationships between soil GHG fluxes and related environmental factors

Soil CO_2 fluxes were more correlated with Ts than Ms at the 10 cm depth. The relationships between soil CO_2 flux and Ts and between soil CO_2 flux and Ms were fitted with exponential growth equations and linear equations, respectively (Fig. 4 and Table 3). Ts and Ms alone explained 89% and 15% of the variability observed in soil CO_2 fluxes at the control, respectively (Table 3). The regression slope between soil CO_2 flux and Ts, as well as Q_{10} value, at the N40 treatment was higher than at the control, and an opposite response was found at other three N treatments (Table 3). All significant continuous variables (Ts, Ms) and categorical variables (N level and N form) could as a whole explain more than 75% of the variation in soil CO_2 fluxes ($p < 0.01$, Table 3). Low and high levels of $NaNO_3$ (N40 and N120) conversely affected soil CO_2 flux (Table 3).

No significant relationships between soil CH_4 flux and Ts were found in any experimental treatments (Fig. 5(a)–(c)). However, soil CH_4 flux was significantly and positively correlated with Ms, and Ms could explain 16%–19% of the variation of soil CH_4 fluxes (Fig. 5(d)–(f) and Table 3). Compared with the control, ammonium-N fertilizer tended to decrease the slopes of regression equation, while nitrate-N fertilizer increased them (Table 3).

Similar to soil CO_2 flux, the relationships between soil N_2O flux and Ts, and between soil N_2O flux and Ms were fitted with exponential growth equations and linear equations, respectively (Fig. 6). Ts and Ms could explain 22% and 31% of variation observed soil N_2O flux at the control (Table 3). Furthermore, the slopes of regression between soil N_2O flux and Ts, as well as Q_{10} values, increased with the levels of N addition (Table 3). Except A120, other three N treatments decreased the slope of linear equation between soil N_2O flux and Ms (Table 3). All significant continuous variables and categorical variables could explain 29% of the variation in soil N_2O fluxes ($p < 0.001$, Table 3). N120 and A120 treatments significantly increased soil N_2O emission (Table 3).

For the cumulative soil GHG fluxes, we found that the annual cumulative CO_2 emission firstly increased then decreased with

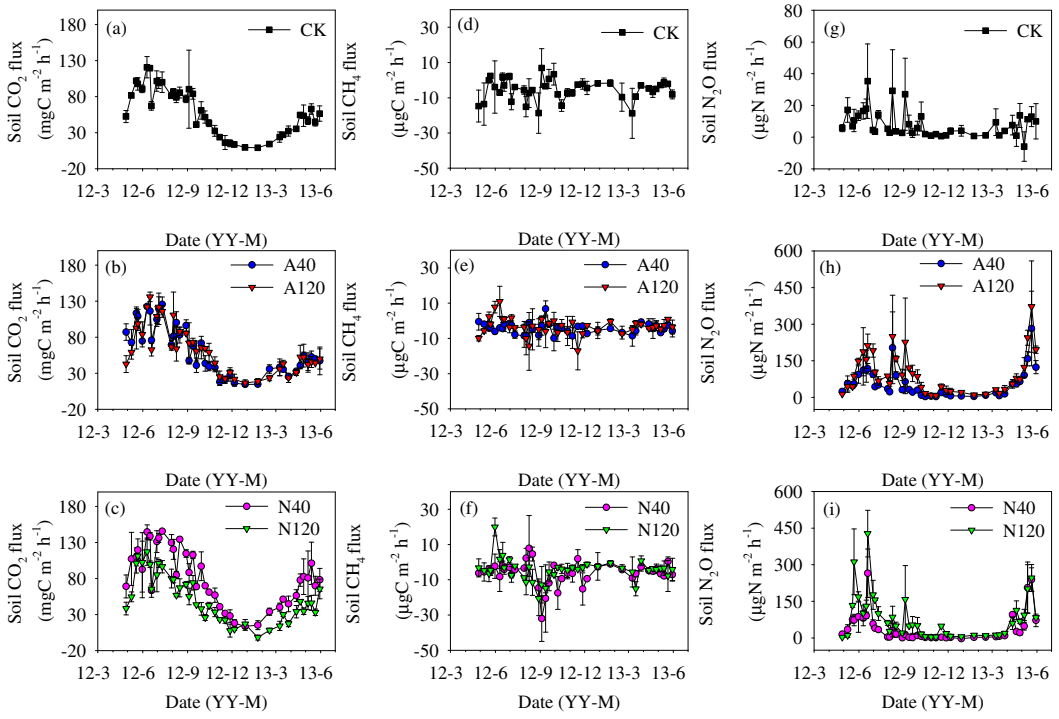


Fig. 3. Monthly variation of soil CO_2, CH_4 and N_2O fluxes under different experimental treatments. Data are shown as means with standard errors ($n = 6$).

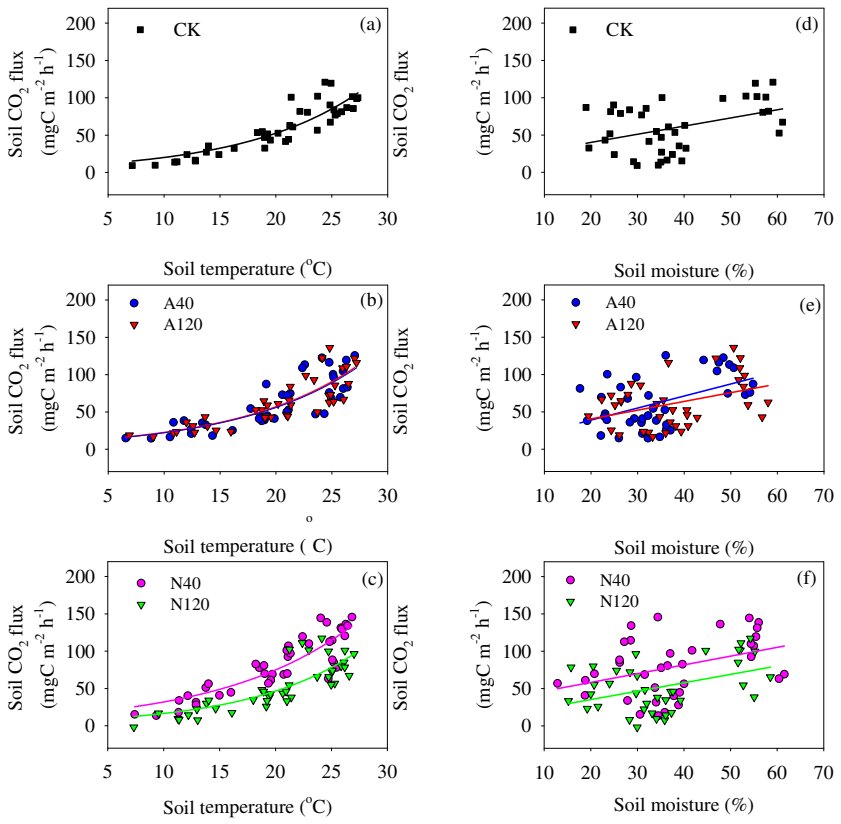


Fig. 4. Relationships between soil CO_2 flux and 10 cm soil temperature, and between soil CO_2 flux and 10 cm soil moisture.

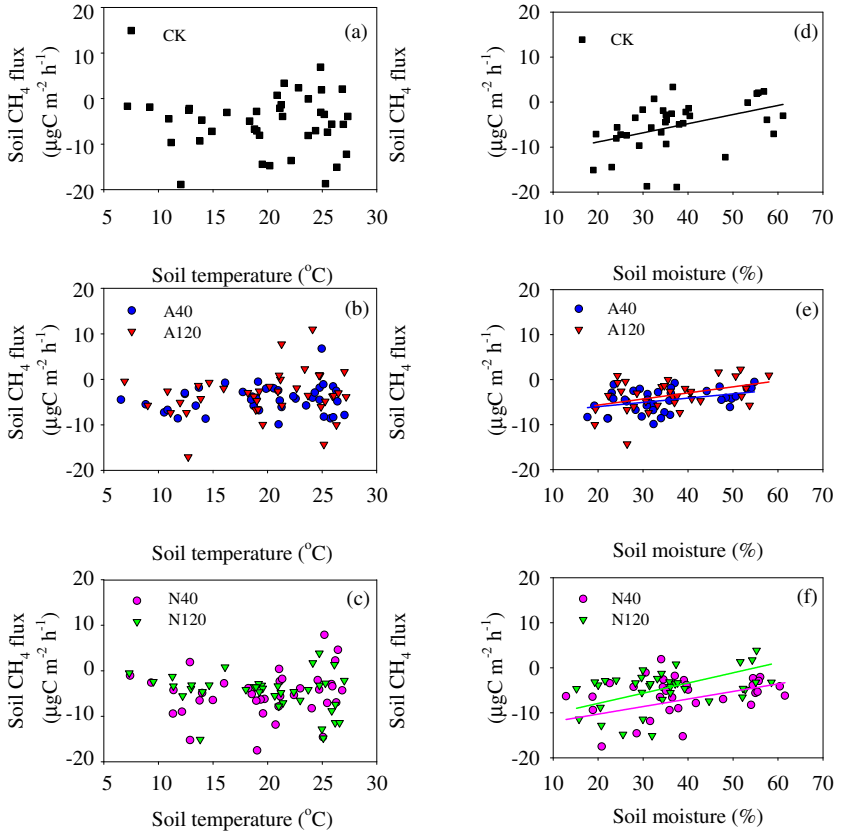


Fig. 5. Relationships between soil CH_4 flux and 10 cm soil temperature, and between soil CH_4 flux and 10 cm soil moisture.

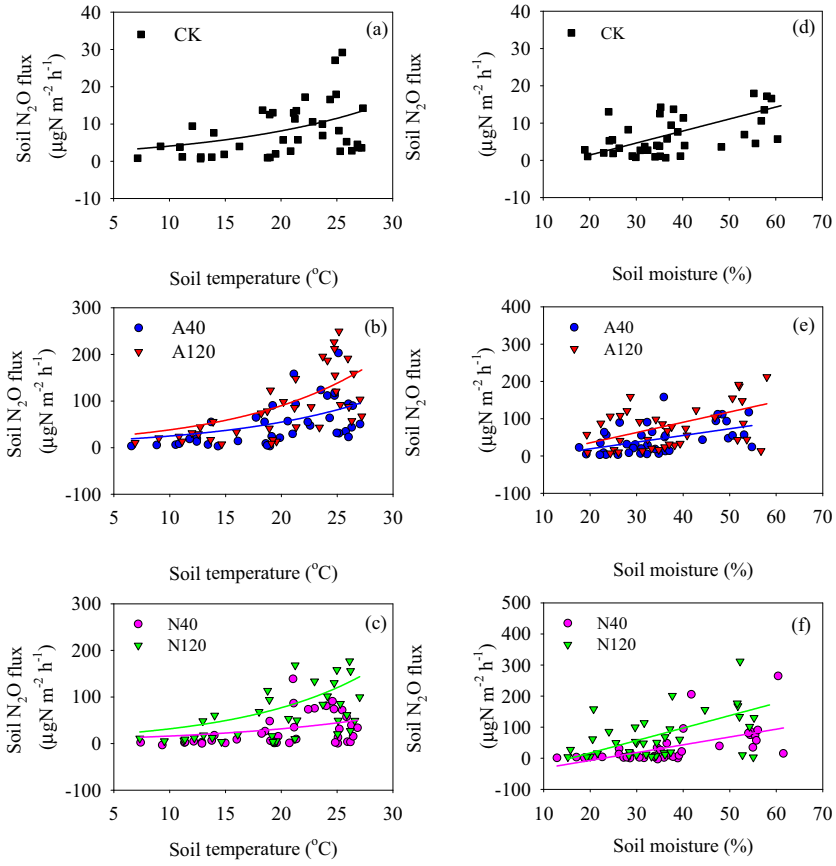


Fig. 6. Relationships between soil N_2O flux and 10 cm soil temperature, and between soil N_2O flux and 10 cm soil moisture.

the level of $NaNO_3$ addition, and the critical load of N inputs was 40 kg N ha^{-1} yr^{-1}; a slight increase in cumulative soil CO_2 emission from low and high levels of NH_4Cl addition was observed (Fig. 7(a)). Also, the cumulative CH_4 uptake decreased with the increasing level of N addition; moreover, the decrease was more significant in A120 than in N120 treatments (Fig. 7(b)). On the contrary, the cumulative N_2O emission increased with the level of N addition, and ammonium-N fertilizer had a stronger promotion than nitrate-N fertilizer (Fig. 7(c)).

4. Discussion

4.1. Effects of simulated N deposition on soil CO_2 emission

The annual cumulative CO_2 emission from the subtropical plantation forest soil averaged 4.66 ± 0.97 t C ha^{-1} yr^{-1}, which was close to those of rubber plantation in southwest China (3.07–3.91 t C ha^{-1} yr^{-1}) (Werner et al., 2006) and the evergreen broadleaved forests (7.7 ± 4.6 t C ha^{-1} yr^{-1}) in southern China (Tang et al., 2006), while substantially lower than those reported from tropical Longan orchards (10.04–18.47 t C ha^{-1} yr^{-1}) (Liu et al., 2008). This could be attributed to the difference in forest type (coniferous vs. broadleaf) and C substrate availability in soils. We found that N addition significantly increased or did not change soil CO_2 emission with the greatest promotion occurring at the low level of $NaNO_3$ addition treatment (Table 2), which indicates the promotion to root autotrophic respiration is greater than or equal to the inhibition to microbial heterotrophic respiration under N enrichment. These results confirm our first hypothesis that increased NO_3^- deposition would promote soil CO_2 emission through increasing root heterotrophic respiration depending on the level of NO_3^- addition. In our study site, root autotrophic respiration and microbial heterotrophic respiration account for 37% and 63% of total soil CO_2 flux, respectively (Wang et al., 2012). The higher promotion of CO_2 flux by NO_3^-–N than by NH_4^+–N could be mainly attributed to the distinct fate of deposited NH_4^+ and NO_3^- in the subtropical plantation. Deposited NH_4^+ is strongly absorbed onto cation exchange sites in soil organic matter and clay minerals, and its accumulation in soils will inhibit the activity of soil microbial community and the decomposition of organic matter (Koba et al., 2003). On the contrary, deposited NO_3^- is very mobile and is absorbed by plants and soil microbes (Inselsbacher et al., 2010). Based on a ^{15}N tracer experiment in the subtropical forests of southern China, Sheng et al. (2014) also reported that more $^{15}NH_4^+$ was recovered from organic and mineral soils, while a large proportion of $^{15}NO_3^-$ was recovered from plant roots. In the early stage of N saturation, most of the deposited N is sequestered within the vegetation biomass (Bowden et al., 2004), and the increased C allocation to root systems could increase autotrophic respiration (Kou et al., 2015).

Except high level of $NaNO_3$ addition treatment, other N addition treatments tended to decrease the temperature sensitivity of soil CO_2 flux (Q_{10}) (Fig. 4(a)–(c)), implying that substrate degradation will decrease with temperature under N enrichment. Nitrogen deposition, to a certain extent, can alleviate the negative effects of warming on soil C cycle. The potential mechanisms can be attributed to the follows: (1) N addition decreases soil respiratory substrates (Chen et al., 2013). The Q_{10} value is negatively correlated with the availability of C in soils (Conant et al., 2011). Elevated N deposition increases both the concentrations of labile organic C (e.g., particulate organic Candreadily oxidizable organic C) and nonreadily oxidizable organic C within the topsoils in the

Table 2
The soil temperature and moisture at 0–10 cm depth, average and cumulative annual fluxes of soil CO_2, CH_4 and N_2O, emission factor of N fertilizer (EF), and total GWP value under different experimental treatments.

Treatment[a]	Ts[b] (°C)	Ms[b] (%)	Average GHG flux[b]			Cumulative GHG flux[b]			EF[b] (%)	Total GWP[b] (t CO2-eq. ha^{-1} yr^{-1})
			CH_4 ($\mu g\,C\,m^{-2}\,h^{-1}$)	CO_2 ($mg\,C\,m^{-2}\,h^{-1}$)	N_2O ($\mu g\,N\,m^{-2}\,h^{-1}$)	CH_4 (kg C ha^{-1} yr^{-1})	CO_2 (t C ha^{-1} yr^{-1})	N_2O (kg N ha^{-1} yr^{-1})		
CK	20.08a (0.89)	38.12a (2.09)	−5.45ab (0.15)	59.53b (0.81)	8.21c (0.21)	−0.56b (0.13)	4.66b (0.97)	0.95d (0.26)	1.00 b (0.00)	4.93b (0.90)
A40	19.65a (0.89)	34.53b (1.76)	−4.36ab (0.077)	61.81b (0.84)	57.00ab (1.49)	−0.46b (0.13)	5.17b (0.39)	4.41b (0.81)	8.67a (1.91)	6.48ab (0.15)
N40	19.94a (0.86)	37.01ab (2.18)	−6.13b (0.17)	81.48a (0.97)	42.00b (1.60)	−0.45b (0.097)	6.86a (0.65)	3.19c (0.84)	5.60ab (2.62)	7.80a (0.78)
A120	19.87a (0.88)	37.23ab (1.85)	−3.23a (0.13)	62.00b (0.81)	96.78a (2.12)	−0.34a (0.078)	5.12b (0.29)	7.48a (2.32)	5.47ab (1.94)	7.34a (0.83)
N120	19.92a (0.85)	35.28b (2.05)	−4.52ab (0.16)	52.34b (0.81)	82.83a (2.31)	−0.39a (0.12)	4.34b (0.10)	5.88a (2.07)	4.10 ab (1.93)	6.08ab (0.63)

[a] CK, A40, N40, A120, and N120 are control (0 kg N ha^{-1} yr^{-1}), low level of NH_4Cl (40 kg N ha^{-1} yr^{-1}), low level of $NaNO_3$ (40 kg N ha^{-1} yr^{-1}), high level of NH_4Cl (120 kg N ha^{-1} yr^{-1}), and high level of $NaNO_3$ (120 kg N ha^{-1} yr^{-1}), respectively.
[b] Data are shown as means with standard errors. Different lowercase letters indicate significant differences among the five treatments.

Fig. 7. Changes of cumulative annual fluxes of soil CO_2, CH_4 and N_2O with the increase in N addition levels.

subtropical forests (Chen et al., 2012), which can partly explain the variation of the Q_{10} value caused by N addition. (2) N addition inhibites litter and soil organic matter decomposition as well as soil CO_2 emission through decresing C-degrading enzyme activities (Conant et al., 2011; Stone et al., 2012) and changing microbial community composition (Zhao et al., 2014). Our recent study also documented that high level of NH_4Cl addition significantly reduced soil fungal biomass (fungal PLFA) and changed microbial community composition (ratio of fungal to bacterial (F/B) PLFAs) (Wang et al., 2015).

According to the theory of N saturation, the promotion of N deposition to soil CO_2 emission is transient, and will shift to the stages of no change and inhibition (Sutton et al., 2011). We also found that N addition at the level of >40 kg N ha^{-1} yr^{-1} significantly inhibited soil CO_2 emission (Fig. 7(a)). The critical level of N input appears to be 70 kg N ha^{-1} yr^{-1} included the ambient level of atmospheric N deposition (about 30 kg N ha^{-1} yr^{-1}, Zhu et al., 2015). However, referring to the N critical loads of forest ecosystems determined for European (Bobbink and Hettelingh, 2011) and North American (Pardo et al., 2011), it seems that the N critical load has been exceeded at the control treatment.

4.2. Effects of simulated N deposition on soil CH_4 uptake

The subtropical plantation forest soils exhibited a sink of atmospheric CH_4 with a mean annual CH4 uptake of 42.3 ± 0.15 $\mu g\,C\,m^{-2}\,h^{-1}$, which is very close to those observed from the subtropical forests of southern China (41.1 $\mu g\,C\,m^{-2}\,h^{-1}$, Zhang et al., 2008b) and the tropical rain forest in Australia (46.67 $\mu g\,C\,m^{-2}\,h^{-1}$, Rowlings et al., 2012). In general, N addition can strongly inhibit CH_4 oxidation in forest soils through the following mechanisms: the competitive inhibition of CH_4

Table 3
The regression results of soil CO_2, CH_4, and N_2O fluxes with soil variations.

Treatment[a]	a	b	c	p	R^2	Q_{10}
$(a)F_{CO_2} = a * exp(b * Ts)$						
CK	4.33	0.12		<0.001	0.89	3.35
A40	7.76	0.097		<0.001	0.77	2.64
N40	8.40	0.11		<0.001	0.84	2.93
A120	8.77	0.091		<0.001	0.81	2.48
N120	3.33	0.13		<0.001	0.78	3.57
$(b)F_{CO_2} = a + b * Ms$						
CK	18.28	1.09		0.010	0.15	
A40	6.79	1.61		<0.001	0.22	
N40	34.72	1.18		0.020	0.12	
A120	17.18	1.61		0.020	0.13	
N120	15.71	1.08		0.010	0.15	
Combined	$F_{CO_2} = 2.27\exp(0.14Ts) + 0.56Ms + 1.62A_{40} - 1.41A_{120} + 17.81N_{40} * -9.48N_{120} * (R^2 = 0.79, p < 0.01)$[a]					
$(c)F_{CH_4} = a + b * Ms)$						
CK	12.90	0.20		0.010	0.16	
A40	−7.88	0.094		0.014	0.16	
N40	−13.66	0.17		0.014	0.16	
A120	−8.35	0.14		0.016	0.17	
N120	−12.45	0.23		0.006	0.19	
$(d)F_{N_2O} = a * exp(b * Ts)$						
CK	0.91	0.082		0.003	0.22	2.27
A40	2.05	0.13		<0.001	0.46	3.82
N40	0.79	0.13		0.014	0.16	3.79
A120	5.23	0.12		<0.001	0.47	3.41
N120	2.09	0.15		<0.001	0.35	4.44
$(e)F_{N_2O} = a + b * Ms$						
CK	−5.02	0.32		<0.001	0.31	
A40	−16.63	1.80		0.002	0.22	
N40	−55.87	2.50		<0.001	0.32	
A120	−3.23	2.23		0.014	0.13	
N120	−63.33	4.01		<0.001	0.27	
Combined	$F_{N_2O} = 0.22\exp(0.19Ts) + 0.53Ms + 12.75A_{40} - 45.28A_{120} * + 2.31N_{40} + 44.95N_{120} * (R^2 = 0.29, p < 0.01)$[a]					

[a] CK, A40, N40, A120, and N120 are control (0 kg N ha^{-1} yr^{-1}), low level of NH_4Cl (40 kg N ha^{-1} yr^{-1}), low level of $NaNO_3$ (40 kg N ha^{-1} yr^{-1}), high level of NH_4Cl (120 kg N ha^{-1} yr^{-1}), and high level of $NaNO_3$ (120 kg N ha^{-1} yr^{-1}), respectively.
Ts and Ms are soil temperature and moisture at 10 cm depth, asterisk "*" means significant difference between N treatment and control.

monooxygenase, toxic inhibition by hydroxylamine (NH_2OH) and nitrite (NO_2^-) produced via NH_4^+ oxidation, and osmotic stress due to high concentrations of NO_3^- and/or NH_4^+ (Bodelier, 2011). In our study, soil CH_4 uptake decreased with the level of N addition (Fig. 7(b)), but one-year N addition did not significantly change the annual cumulative CH_4 uptake (Table 2). The contrasting effects between NH_4^+ and NO_3^- were not appeared, which was inconsistent with our second hypothesis. A proposed explanation is that one-year N addition did not significantly cumulate soil NH_4^+–N in our study (Wang et al., 2014), and the available N concentration in the ecosystem did not meet the threshold of atmospheric N deposition. Similar results including promotion and no change are widely seen in the boreal and temperate forests (Whalen and Reeburgh, 2000; Saari et al., 2004; Maljanen et al., 2006; Xu et al., 2014). Furthermore, soil CH_4 uptake is mainly driven by soil moisture (Fig. 5), and the decrease in soil moisture could promote CH_4 oxidation in soil profile (Table 2). This promotion could offset the inhibition caused by inorganic N inputs (Fang et al., 2014).

4.3. Effects of simulated N deposition on soil N_2O emission

Annual cumulative soil N_2O flux under nature condition averaged 0.95 ± 0.21 kg N ha^{-1} yr^{-1}, which is less than those of the subtropical forests in southern China (3.03–3.20 kg N ha^{-1} yr^{-1}) (Tang et al., 2006; Liu et al., 2008). This is attributed to the difference in soil N contents among the sites (Tang et al., 2006; Liu et al., 2008; Wang et al., 2014). Ammonium-N and nitrate-N addition significantly increased cumulative annual soil N_2O emission by 5.2 and 3.8 folds, respectively (Table 2 and Fig. 7(c)). This result confirms our second hypothesis that the promotion to soil N_2O emission was greater from deposited NH_4^+ deposition than from deposited NO_3^-. In the subtropical forests of southern China, only 4.1–8.67% of the total N_2O emissions were derived from experimental N deposition (Table 2), which is close to the estimated ranges in the temperate beech and spruce forest of Europea (6–13%, Eickenscheidt et al., 2011). Also, simulated NH_4^+ and NO_3^- deposition increased the temperature sensitivity of soil N_2O flux (Fig. 6), which is mainly attributed to the increase in soil inorganic N substrates and ammonia-oxidizing microbes (Isobe et al., 2012; Wang et al., 2014). Although some evidences suggest that soil denitrification dominates the production and emission of soil N_2O (Zhang et al., 2009, 2012a), so far we cannot quantify the relative contributions of nitrification in upper soil layers and denitrification in deeper soil horizons to the cumulation and emission of N_2O in soils. Nitrogen and oxygen isotope tracer should be a good technique to solve the question in future studies.

5. Conclusions

A complete perspective was proposed in this study to assess the effects of deposited NH_4^+ and NO_3^- ions on soil GHG fluxes and total GWP. Our findings demonstrated that short-term N addition promoted soil CO_2 and N_2O emissions, but did not change soil CH_4 uptake in the subtropical plantation, southern China. Ammonium-N fertilizer addition had a greater promotion to soil N_2O emission than nitrate-N fertilizer addition. In contrary, the promotion to soil CO_2 emission was greater from nitrate-N fertilizer addition than from the ammonium-N addition. Overall, elevated atmospheric N deposition changed the temperature sensitivity of soil CO_2 and N_2O emissions and would exacerbate global warming. Therefore, it is necessary to evaluate the combined effects of atmospheric N deposition on carbon sequestration and GHG emission in the subtropical plantation. We recommend that nitrate-N fertilizer should

be applied to increase ecosystem productivity and to mitigate soil GHG emission in the subtropical plantation, southern China.

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