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Biochar addition can reduce NO_x gas emissions from a calcareous soil

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ABSTRACT

The effect of biochar (BC) addition on NO_x gas emissions was investigated from a calcareous soil. Application rates of zero (BC0), 1 (BC1), 2 (BC2), 5 (BC5), and 10 (BC10) percent weight/weight were used. The NO_x emission flux was measured in a dynamic flux chamber. The flux was generally higher in BC0 than in the amended plots. The total NO_x emission from BC1, BC2, BC5, and BC10 fell by 50.3, 75.3, 80.4, and 79.6%, respectively, relative to BC0. The emission flux from the BC5 showed a minimum average of $21.1 \pm 13.5 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO, $-0.81 \pm 1.31 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO₂, and $20.6 \pm 13.8 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO_x. By comparison, the measured maximum average emission flux from the BC0 was $107.2 \pm 30.98 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO, $-2.31 \pm 2.56 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO₂, and $105.3 \pm 45.3 \mu\text{g N}/(\text{m}^2 \cdot \text{h})$ for NO_x. The results indicate that biochar amendment can potentially reduce NO_x emissions.

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Introduction

The application of nitrogen fertilizer to soil is key to promoting and sustaining agricultural production [1]. A significant portion of fertilizer N is lost by leaching [2], gas emissions [3,4] and soil erosion [5]. Such losses have a negative effect on farmers' income as well as on the immediate environment.

Gaseous N, in the form of NH₃, NO, N₂O and dinitrogen (N₂) can be lost by volatilization, nitrification, and denitrification. Nitrogen oxides (NO_x) gas, comprising NO and NO₂, play a key role in the photochemical synthesis of tropospheric ozone (O₃) [6]. Since the emission of NO_x from soil is largely mediated by soil microbes, the process is subject to such environmental variables as soil pH, soil water content, temperature, and the availability of inorganic nitrogen [7,8].

Recent studies indicate that the soil NO_x flux can be on a par with that from fossil fuel combustion [9–11]. This is especially the case in rural areas, where the agricultural soils emission often dominates the total flux [12–14]. Therefore, it is imperative to find a low-cost and high-efficiency method for reducing soil N loss.

Biochar is produced by heating carbonaceous organic matter under oxygen-limited conditions [15,16]. Because of its great surface area, high micro-porosity and other physiochemical properties depending on the biomass feedstock and the pyrolysis conditions [17–20], biochar is very effective in sorption nutrient elements in soil. Biochar

amendment to soil increases N retention [21], enhances N use efficiency [22] and promotes agricultural production [23,24], recovers excess N [25], and reduces N leaching [26,27]. Furthermore, the addition of biochar input to the soil would be expected to affect soil nitrification and denitrification process controlling the release of soil nitrogen gas [28,29].

Biochar application to soil for carbon sequestration as well as agricultural and environmental sustainability has attracted much interest [30–33]. Little is known, however, about the effects of biochar addition on NO_x emission [26]. Furthermore, the focus of research has been directed on highly weathered soils of the humid tropics, acidic forest soils [34,35], mollisol [36], temperate soil [37] and agricultural soil [38]. On the other hand, the emission of greenhouse gases, such as CO₂, CH₄, N₂O and NH₃ under both laboratory and field conditions is well documented although the results have not always been consistent [28,29,39–43]. However, the factors controlling trace gas emissions from biochar-amended soils are not well understood, especially with respect to calcareous agricultural soils [44–46]. Therefore, understanding the impact of biochar addition on the flux of trace gases in agricultural soils is of great significance for the application of biochar. To test the hypothesis that biochar addition to calcite-rich soils can reduce NO_x emission, we have measured the fluxes of NO_x from a calcareous sandy loam in Southwest China. By

varying the rate of biochar application, we would obtain an insight into the factors controlling NO_x emissions, and deduce an optimum rate of amendment.

Materials and methods

Field experiments site and biochar preparation

The field experiments were carried out on a calcareous sandy loam (N 27°0'40.20", E 107°2'46.95", 1118 m) located in the karst region of Kaiyang County, Guiyang City, Guizhou Province, Southwest China (Figure 1(a)). The upland site has a mean annual rainfall of 926–1419 mm and a mean annual temperature of 10.6–15.3°C for the last 30 years. Some properties of the soil are listed in Table 1.

The biochar was obtained by pyrolysis at 550°C of crop stalks, mainly composed of rapeseed (*Brassica campestris* L.) and maize (*Zea mays* L.) straw. The biochar produced has a pH of 10.3 ± 0.2, and its C, N, NH₄⁺ and NO₃⁻ contents are 80% ± 3%, 1.2% ± 0.05%, 1.0 ± 0.02 mg^okg⁻¹ and 8.6 ± 0.3 mg^okg⁻¹, respectively. The calcareous sandy loam has a pH of 8.23 ± 0.21, while its C, N, NO₃⁻ and NH₄⁺ contents are 4.3% ± 0.1, 0.2% ± 0.01, 5.26 ± 0.43 mg^okg⁻¹ and 3.67 ± 0.17 mg^okg⁻¹, respectively.

Experimental design

The area of each experimental plot was 2 × 4 m². A total of five treatments were used, each of which had three replicates: (1), plot without biochar (control plot); (2), plot amended with biochar at a rate of 1% (w/w); (3), plot amended with biochar at a rate of 2% (w/w); (4), plot amended with biochar at a rate of 5% (w/w); and (5), plot amended with biochar at a rate of 10% (w/w).

The selected field plots have been planted with maize (*Zea mays* L.), rapeseed (*Brassica napus* L.), and vegetables for more than 10 years, being cultivated and managed by local farmers using traditional methods. Water is supplied by atmospheric precipitation, while the cropping system is one of rotating rapeseed and maize (rapeseed-maize).

The field experiment was done by mixing the top 10 cm soil with biochar at an application rate of 1, 2, 5, and 10% w/w in July 2010 prior to planting maize. The NO_x emission flux was monitored from 14 September, 2010 to 7 September, 2011. The field gas sampling was performed once every 7 or 20 days depending on the season. NO_x flux was measured from 11:00 a.m. to 6:00 p.m. The sampling time was chosen because fluxes at the stated times approached the average of five diurnal measurements (from 7:00 a.m. to 7:00 p.m.). Table 2 shows specific crops, fertilizer types and amounts applied.

Determination of soil variables

Soil samples were taken from the top 10 cm of the soil profile near the chamber using a cylinder ring sampler (100 cm³). Soil bulk density for the 0–10 cm depth (n = 10) was determined using the core method. Soil pH was obtained by mixing air-dried soil with deionized water at a 1:5 (w/w) ratio, and measuring with a pH meter. Soil water content was calculated from the difference in weight between the fresh and oven-dried (105°C) soil, divided by the oven-dried soil weight. The contents of ammonium (NH₄⁺) and nitrate (NO₃⁻) were obtained by extracting the soil with 2 mol L⁻¹ KCl, and measuring the extracts by colorimetry [47]. The determination of soil organic matter was based on the Walkley-Black chromic acid wet oxidation method [48]. Total carbon (TC) and total nitrogen (TN) contents of the soil were determined with an elemental analyzer (PE2400-II, Perkin, American).

Measurements of NO_x fluxes

NO_x fluxes were measured using a dynamic flux chamber (DFC) method similar to that used previously by Wang et al [8] (Figure 1(b)). Net fluxes from the soils were obtained by comparing the flux in the sampling chamber with that in the reference chamber. NO_x fluxes (F), expressed as μg N/(m²·h), were calculated using the following equation:

$$F = (C - C_0) \times Q/A \quad (1)$$

where C (μg/m³) is the NO_x concentration in the outflow air of the chamber, C₀ is the NO_x concentration in the inflow air (μg/m³), which is drawn at the height 1.5 m above the ground, Q is the flow rate of the inflow air (m³/h), and A (m²) is the area covered by the chamber. C was determined using the average concentration measured during a 20-min period after the NO_x concentration had reached a dynamic equilibrium in the chamber.

Statistical analysis

The statistical software package SPSS 19.0 (Chicago, IL, USA) was used for descriptive statistics, One-way ANOVA, linear regression, correlation analysis and paired sample T-test of NO_x fluxes in different treatments. Descriptive statistics were used to describe the basic features of the soil properties after biochar addition. One-way ANOVA was used to test the difference between multiple means, and determine whether the different rates of biochar application have a significant influence on the release of NO_x gas. Linear regression and correlation analysis were used to correlate different treatments with soil

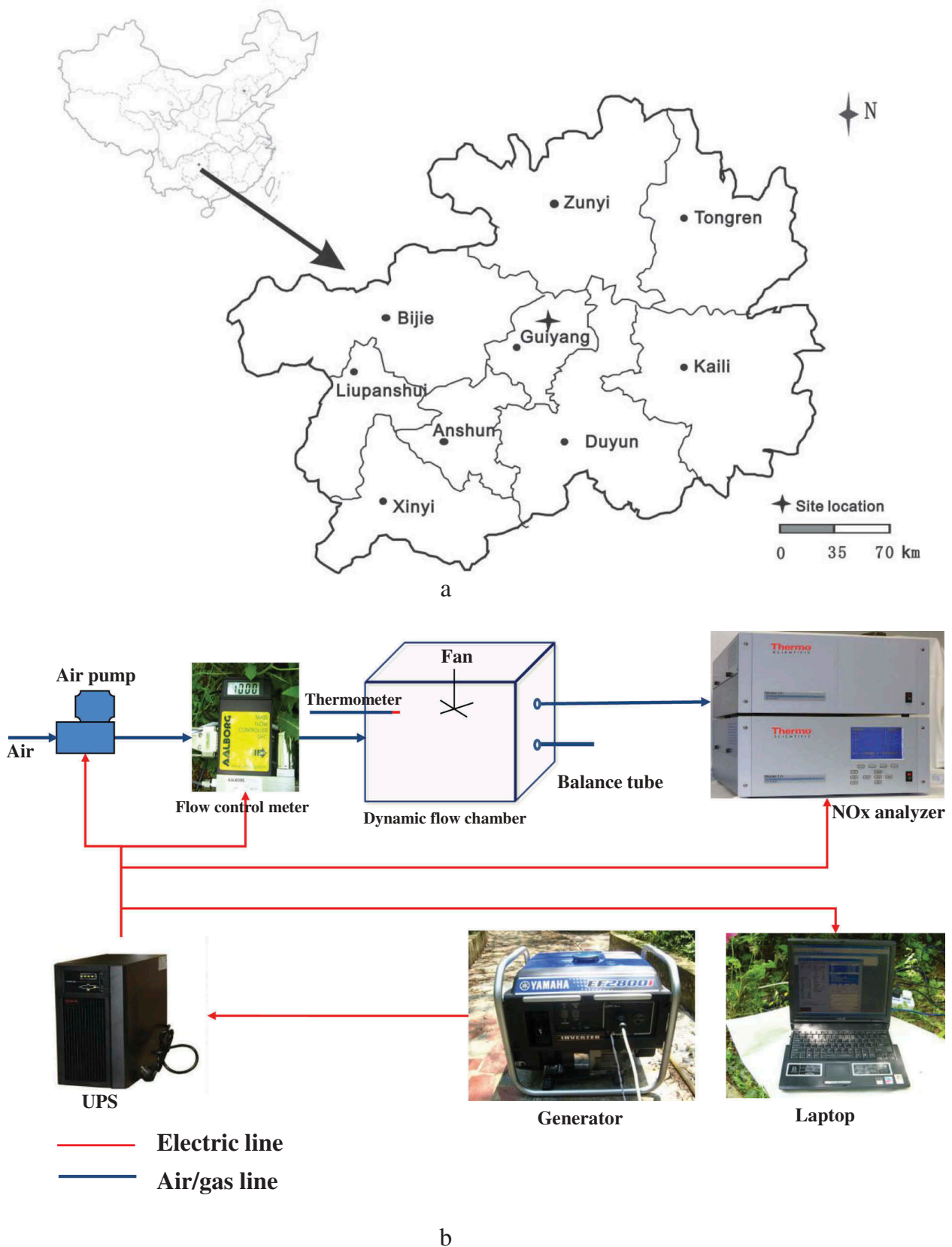


Figure 1. Map of the location of the study (a) and field observation experimental design (b).

properties. Paired sample T-test was used to analyze the difference between BC0 and BC1, BC2, BC5 and BC10 in terms of NO and NO_x emission fluxes. The statistical software package SigmaPlot 12.5 (SAS, Cary, NC) was used for creating statistical graphs.

Results and discussion

Biochar effect on soil properties

In agreement with previous measurements [8], the correlation between NO_x fluxes and soil temperature was significant ($P < 0.05$) for all five plots (Table 3).

Table 1. Some soil properties before and after biochar addition.

Soil characteristics	BC0	BC1	BC2	BC5	BC10
Bulk density (g cm ⁻³)	1.32a	1.24b	1.21c	1.10d	0.96e
pH	8.23a	8.27a	8.36a	8.49b	8.57b
NH ⁺ ₄ -N, mg kg ⁻¹	3.67a	3.72a	5.55b	4.49c	4.46c
NO ⁻ ₃ -N, mg kg ⁻¹	5.26a	7.56b	7.90b	9.04c	15.06d
Soil organic matter (%)	3.14a	5.25b	7.33c	8.75d	14.50e
Total carbon (g kg ⁻¹)	49.6a	55.3b	61.9c	90.4d	108.0e
Total nitrogen (g kg ⁻¹)	2.67a	2.48a	2.57a	3.83b	4.00b

Means in the same row with different letters were significantly different ($P < 0.05$, ANOVA, LSD) with treatments as the fixed variable ($n = 3$).

This finding may be explained in terms of the effect of biochar addition on nitrification, denitrification, and NO_x gas emissions [42,49,50]. These microbially-controlled processes are influenced by many factors, including soil water content, temperature, ammonium and nitrate concentrations and pH [51]. Among them, soil temperature is considered to be of major importance because of its effect on microbial activity and gas diffusion rates [8,11]. With an increase in biochar application rate, the soil pH is not significantly different between BC0, BC1 and BC2 treatment. However, there is a significant difference when biochar addition ratio is high in BC5 and BC10 (Table 1). On the other hand, soil bulk density decreases as the biochar application rate increases. As a result, we would expect soil compactness to decrease and soil aeration to increase. Table 1 also shows that the total carbon and total nitrogen contents of the soil increase with biochar application rate.

As shown in Figure 2, soil water content changes from 22.3% for the BC0 (control) plot in August to 54.7% for the BC10 plot in December. Although water content can fluctuate greatly between seasons, the addition of biochar leads to a significant ($P < 0.01$) increase in water holding capacity (WHC). This observation is related to the high capacity of biochar for retaining water in its microporous structure [40].

Furthermore, the presence of hydrophilic groups in, and the extensive surface area of, biochar are conducive to water uptake and retention [52]. Because of its small particle size, biochar may further cause the partial or total blockage of soil pores, decreasing water infiltration while increasing WHC [53].

Figure 3 shows the fluctuation in atmospheric precipitation and temperature in soil (to 5cm depth), surface, chamber, and atmosphere over the experimental period. The variation in soil NO₃⁻ and NH₄⁺ concentrations for the different plots during the same period shows a similar trend (Figure 4). These observations indicate that temperature is the key factor since temperatures in summer are higher than those measured in other seasons. Biochar addition increased the average soil NO₃⁻-N content by 43.7, 50.2, 71.9 and 186 in the BC1, BC2, BC5, and BC10 plots, respectively. On the other hand, soil NH₄⁺ concentrations first increased and then decreased as the rate of biochar application increased. The correlation between biochar addition and NH₄⁺ concentration, however, was not significant ($P > 0.05$). Furthermore, there was a significant increase ($P < 0.01$) in soil water content with a rise in the rate of biochar application (Figure 2).

There is also a good correspondence between mineral N concentration and urea application (Figure 4 and Table 2). The highest NH₄⁺-N content (27.7 mg°kg⁻¹ soil) in the control plot was measured in July 2011 following maize planting and urea application, presumably as a result of urea hydrolysis. Earlier, Rochette et al have reported that subsurface banding of urea can greatly enhance ammonia (NH₃) emission due to a localized increase in soil NH₄⁺ concentration and pH [54]. The maximum NO₃⁻ concentration (116.5°µg g⁻¹ soil) was measured for the BC10 plot in August 2011 indicating a lag with respect to soil ammonium nitrogen. This observation suggests biochar addition promotes nitrification.

Table 2. Specific field crops, types and fertilization events according to the local cultivation habits.

Crops	Date	Fertilizer types	Fertilizer amount (kg)	Farming & growing events
Rapeseed	2010-11-18	Urea	1.0	Planting rapeseed
		Calcium superphosphate	5.5	
		Compound fertilizer	5.5	
		Potash fertilizer	0.5	
		Urea	3.0	
Maize	2011-02-05	Urea	3.0	Planting maize
	2011-02-24	Urea	2.0	
	2011-06-15	Coal fly ash and slag	2.0	
		Compound fertilizer	15.0	
	2011-08-10	Urea	3.25	

Table 3. Spearman correlation coefficients between soil NO_x gas fluxes and soil temperature (in 5 cm depth), soil water content, NO₃⁻ and NH₄⁺.

Treatment	Temperature (°C)	Soil water content (%)	NO ₃ ⁻ (mg kg ⁻¹)	NH ₄ ⁺ (mg kg ⁻¹)
BC0	0.626*	-0.336	0.666*	0.358
BC1	0.610*	-0.427	0.511	0.374
BC2	0.459*	0.114	0.591*	0.336
BC5	0.569*	-0.291	0.595	0.375
BC10	0.593*	-0.164	0.479*	0.054

* $P < 0.05$ (two-tailed). All measurements from the monitoring period are included ($n = 20$ in each treatment).

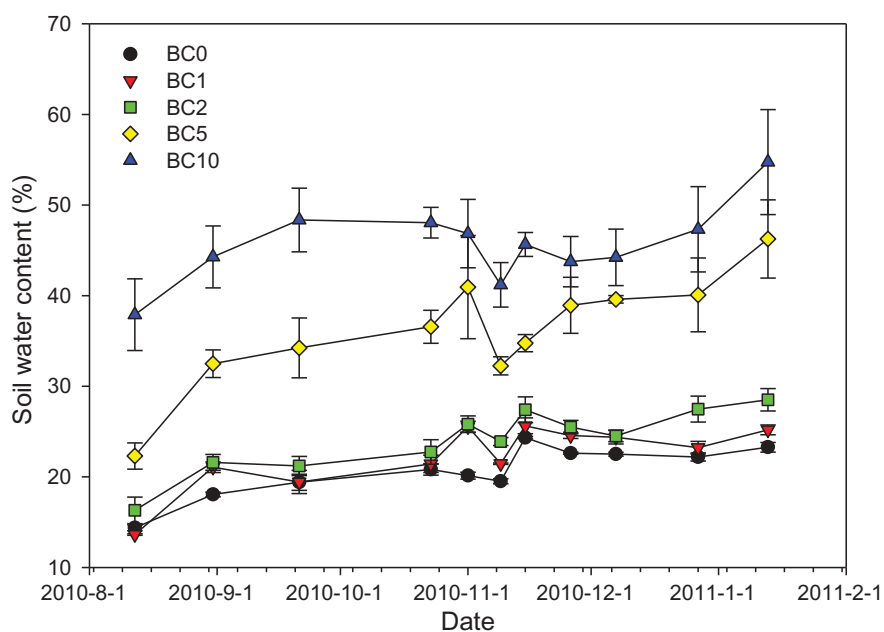


Figure 2. Soil water contents at varying biochar application rates. Paired sample T-test statistical analysis of different treatments revealed that there was a significant difference in each category ($P < 0.01$).

The capacity of biochar for sorbing soil mineral nitrogen while reducing nitrogen dissolution and migration, in dependence of feedstock and pyrolysis conditions, has been the subject of many investigations [23,55–60]. Besides improving soil aeration, biochar application reduces anaerobic microbial activity and inhibits denitrification [61]. By increasing soil pH, biochar addition also enhances the activity of NO-reducing enzymes as well as N_2 formation and ammonia volatilization [62].

Effects of biochar and n fertilizer on NOx emissions

The variation in NO, NO_2 , and NOx emission fluxes from different plots were illustrated in Figure 5, while the range and average value of these emissions are tabulated in Table 4. The minimum average emission of NO ($21.1 \pm 13.5^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$), NO_2 ($-0.81 \pm 1.31^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$), and NOx ($20.6 \pm 13.8^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$) was measured in the BC5 plot. The maximum average emission of NO ($107.2 \pm 30.98^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$), NO_2 ($-2.31 \pm 2.56^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$), and NOx ($105.3 \pm 45.3^\circ\mu\text{g N}/(\text{m}^2\cdot\text{h})$) was measured in the BC0 (control) plot following N fertilization. Addition of biochar decreased NOx emissions compared to the BC0 plot (Table 4). Biochar addition depressed NOx emissions by 50.3% to 80.4%, depending on annual application rate. One-way ANOVA analysis (LSD) indicates a significant difference ($P < 0.05$) in NO and NOx emission fluxes between BC0 and the treated plots except for BC1.

The interactions between biochar, N fertilizer, greenhouse gas emissions, and soil type have been reported by several investigators [39,63,64]. Application of N fertilizer

causes an initial sharp increase in NO_3^- and NH_4^+ as well as NOx emissions [63]. Higher nitrogen oxide gas emissions have been reported with urea- and ammonium-based fertilizers than with nitrate-based counterparts under unsaturated soil conditions [65]. To elucidate the influence of NO_3^- and NH_4^+ on NOx emissions, the correlations between NOx fluxes and NO_3^- and NH_4^+ were assessed for all plots. In the present investigation the correlation between NOx fluxes and soil NO_3^- was significant ($P < 0.05$) but no significant difference was observed between NOx fluxes and NH_4^+ (Table 3). Although NOx emission flux peaked during the fertilization in February 2011, there was no change in soil mineral nitrogen content. This finding may be ascribed to NOx emission or NH_3 volatilization [66,67]. Because the soil is alkaline soil ($\text{pH} > 8.0$), most of the fertilizer N may not enter into the soil after application and probably emission to the atmosphere as a gaseous loss in a short period. At pH levels of 8.0 or above, volatilization of ammonia gas can cause large losses of nitrogen. Several studies have reported the short-lived peaks in NOx flux following fertilization [63,68]. In an early study, Fenn and Kissel found that ammonia was rapidly volatilized from NH_4 -N salts applied to the surface of a calcareous soil [69]. An increase in pH at the soil surface following biochar addition would also induce NH_3 volatilization. Such a process is consistent with the finding that significant NOx emission occurred after fertilizer application although there was no change in soil mineral nitrogen.

The process of nitrogen transformation in the soil can be expressed in terms of the hydrolysis of urea (step 1; Equation (2)) followed by ammonification (step 2; Equation (3)) and nitrification (step 3; Equations (4) and (5)):

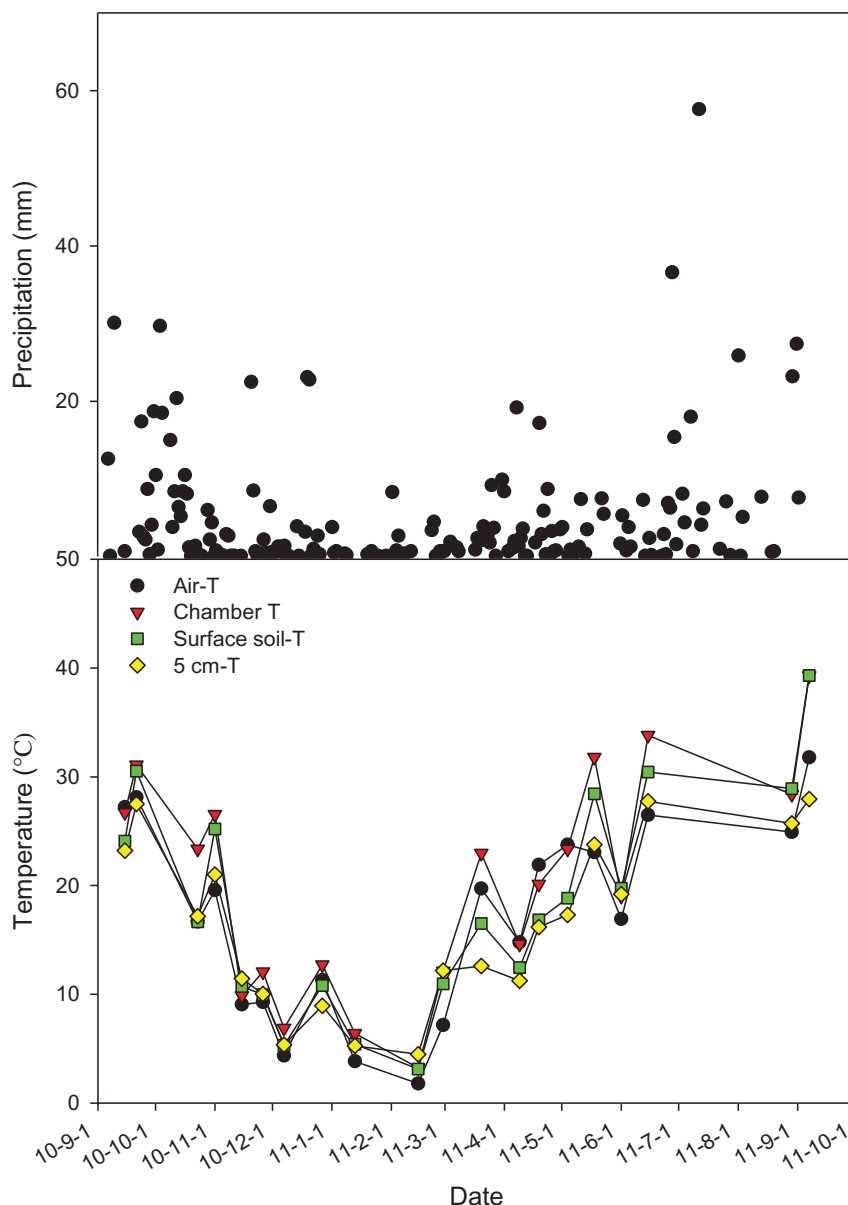
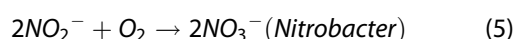
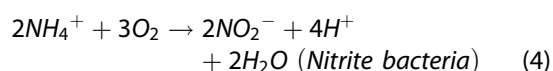
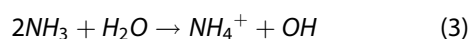
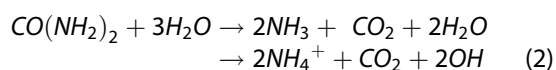


Figure 3. Fluctuation of temperature and rainfall during the observation period.



The appearance of two NO_x emission peaks in March and September 2011 is apparently related to two fertilization events during this period (Table 2). This observation accords with previous findings [63,69–71] that nitrogen fertilizer (urea) addition promotes rapid but short-lived emissions of NO_x [72]. The NO_x flux emission was high for the control plot and low for the BC5 plot, indicating that biochar can

immobilize soil mineral nitrogen [42] and suppress N₂O emissions [73].

The following mechanisms have been proposed to account for the decrease of NO_x emissions: (1), the addition of biochar adsorbs NH₄⁺ and NO₃⁻ which leads to a reduction of the soil inorganic-N pool [74]; (2), The electron-accepting sites on the surface of biochar can increase the redox potential of the soil while reducing the NO source capacity of the soil, and (3), The microorganisms immobilize the available soil N while inhibiting denitrification, thereby reducing the NO source capacity of the soil, and (4), sorption of soil NO, N₂O, and CH₄ by biochar

Correlation between biochar application rate and NO_x emissions

Different application rates of biochar could result in different aerobic and/or anaerobic conditions. Increased soil

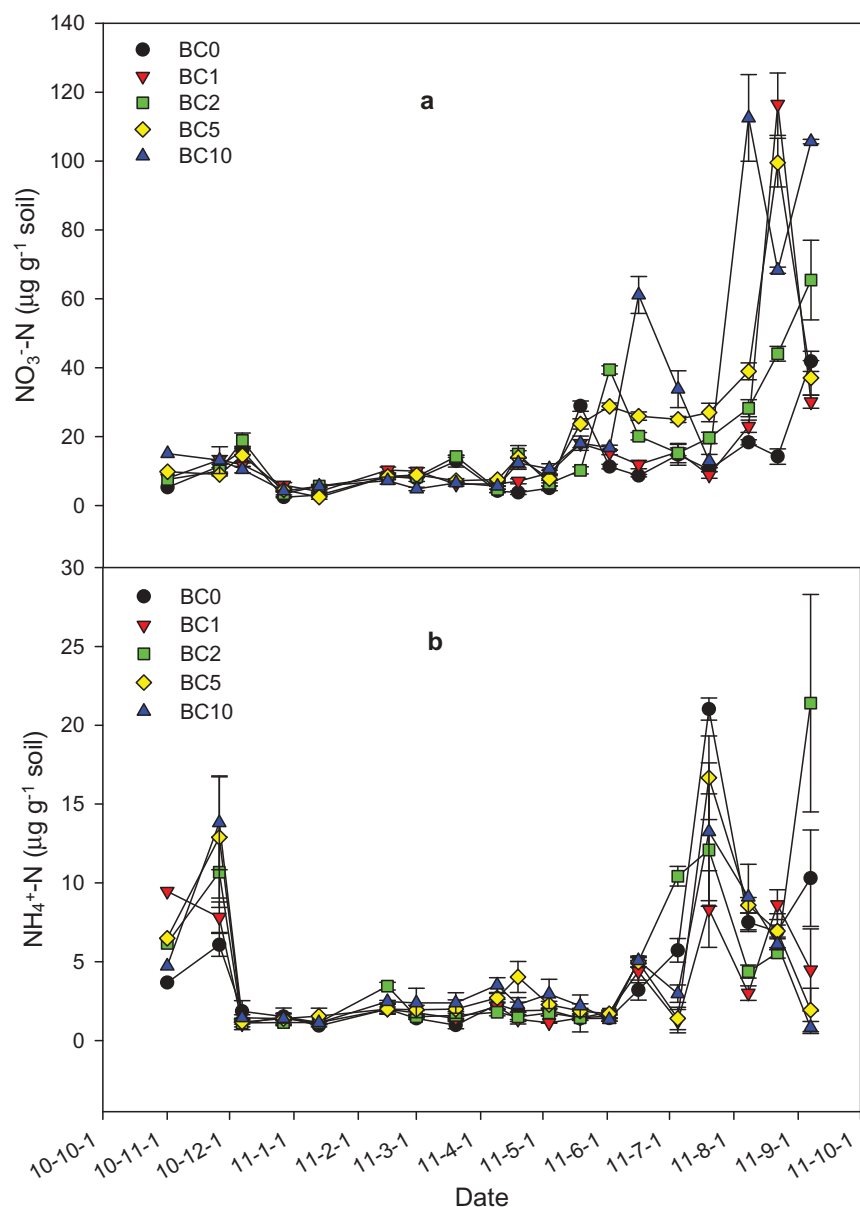


Figure 4. Variations of soil NO_3^- (a) and NH_4^+ (b) at different plots during the whole measurement period. Symbols may cover error bars. Paired sample T-test statistical analysis revealed that there was a significant difference between BC0 and any other treatment in NO_3^- ($P < 0.01$), but there was no significant difference between BC0 and other treatment in NH_4^+ .

aeration from biochar addition could reduce denitrification. Dry aerobic conditions are conducive to nitrification, while anaerobic conditions promote denitrification [75]. However, there is a noted shortage of data about the dose-response of biochar application to soil lacks systematic assessment [45,76,77]. Within a certain range of application rates, a change in soil properties can mitigate NO_x emissions by increasing nitrogen immobilization. Spokas et al have reported that the addition of 2–10% w/w biochar to soil at field capacity can suppress net N_2O production but the data were not a statistically significant [19]. In the present instance, the addition of 2, 5, and 10% w/w biochar led to a significant reduction ($P < 0.05$) in NO_x emission rates. In this regard, the BC5 plot gave the optimum ($P < 0.05$) cost-effective result from economic point of view.

For calcareous soils, a lower or higher application rate is therefore not recommendable. Application rates higher than 5% w/w would increase soil pH and NH_3 gas emission, while a lower rate would not provide enough biochar to immobilize soil mineral nitrogen. This suggestion accords with the observation that the curves relating application rates to average NO_x emission fluxes show a declining trend (Table 4). In addition, the priming effect of fertilizer application without biochar would be more pronounced than in the presence of biochar [78,79]. That the application of 5% w/w biochar to agricultural calcareous soils could significantly reduce NO_x emission fluxes is in keeping with the results of studies regarding the biological response of, and N_2O emissions from, sandy soils treated with biochar [61,80].

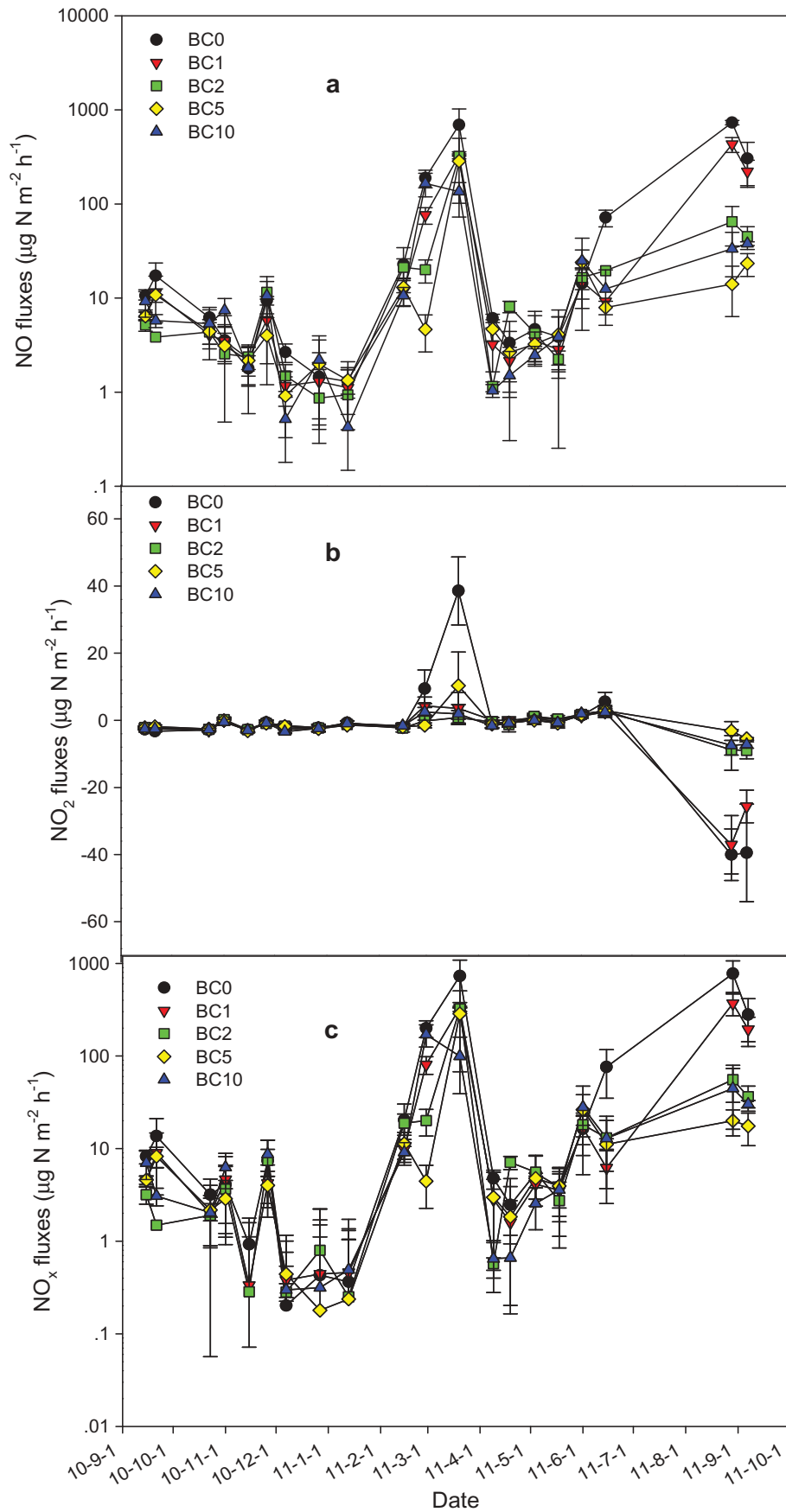


Figure 5. Variations in NO (a), NO₂ (b) and NO_x (c) fluxes at different plots during the whole measurement period. Symbols may cover error bars. Paired sample T-test statistical analysis indicated that there was a significant difference between BC0 and BC1, BC2, BC5 and BC10 in NO and NO_x emission fluxes ($P < 0.05$).

Table 4. Variations of NO/NO₂/NO_x emission fluxes in different treatments.

Treatment	NO ($\mu\text{g N m}^{-2} \text{h}^{-1}$)		NO ₂ ($\mu\text{g N m}^{-2} \text{h}^{-1}$)		NO _x ($\mu\text{g N m}^{-2} \text{h}^{-1}$)	
	Range	Average	Range	Average	Range	Average
Control	1.30 ~ 693	107.2 ± 30.98a	-40.0 ~ 38.5	-2.31 ± 2.56a	0.202 ~ 775	105.3 ± 45.3a
BC1	1.11 ~ 431	57.1 ± 10.0ab	-37.1 ~ 4.25	-3.37 ± 1.62a	0.334 ~ 369	52.3 ± 11.1ab
BC2	0.868 ~ 322	27.9 ± 4.61b	-8.94 ~ 2.90	-1.53 ± 1.15a	0.251 ~ 322	26.0 ± 5.0b
BC5	0.913 ~ 286	21.1 ± 13.5b	-5.47 ~ 10.3	-0.81 ± 1.31a	-0.853 ~ 286	20.6 ± 13.8b
BC10	0.425 ~ 166	23.7 ± 6.81b	-7.42 ~ 2.50	-1.45 ± 0.99a	-1.00 ~ 171	21.5 ± 8.8b

Means in the same column with different letters are significantly different ($P < 0.05$, ANOVA, LSD) with treatments as the fixed variable ($n = 20$ in each treatment).

Conclusions

The present investigation indicates that biochar addition to subtropical calcareous soils can reduce NO_x emissions, and increase soil pH, nitrate N, and water holding capacity but decrease soil bulk density. Fertilization is the main factors affecting the NO_x emissions, overwhelming the effects of temperature and biochar application rates. Consider the application of biochar from economic point of view, applying of 5% w/w biochar is optimum for the calcareous agricultural soil in Southwest China. Because of its alkaline reaction, the soil can lose some of the applied fertilizer N through gaseous NH₃ emission. Biochar addition can potentially mitigate N losses within agroecosystems. Future studies should focus on the relationship between microbial activity and NO_x gas emission in a range of soils.

Disclosure statement

No potential conflict of interest was reported by the authors.

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