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#### **RESEARCH ARTICLE**

# Effects of biochar application on fluxes of three biogenic greenhouse gases: a meta-analysis

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**Abstract.** Biochar application to cropland has been recommended as a strategy to reduce increasing at mospheric CO<sub>2</sub> concentrations and mitigate climate change. However, the direction and magnitude of responses of greenhouse gas (GHG) fluxes to biochar application to cropland remain unclear. Our metaanalysis of 296 observations across 61 studies for the first time quantitatively estimated the effects of biochar amendment on fluxes of three GHGsCO<sub>2</sub>, N<sub>2</sub>O, and CH<sub>4</sub>. The results showed that biochar application led to a significant change in soil GHGs emissions: in general, 19% for CO<sub>2</sub>, -16% for N<sub>2</sub>O (P < 0.05), but no pronounced change in CH<sub>4</sub> emissions; in paddy, -5% for CO<sub>2</sub>, -20% for N<sub>2</sub>O, but +19% for CH<sub>4</sub> (P < 0.05); in upland, -18% for N<sub>2</sub>O, +12% for CO<sub>2</sub>, and high uncertainty for CH<sub>4</sub>. The responses of soil GHG fluxes to biochar application were regulated mainly by experiment length, biochar application rate, biochar properties, providing a new perspective for more comprehensive understanding on biochar. The biochar derived from husk was recommended to apply to cropland with an application rate of 20–30 t·ha<sup>-1</sup>.

**Key words:** CH<sub>4</sub>; charcoal; climate change; CO<sub>2</sub>; CO<sub>2</sub>-C equivalent; N<sub>2</sub>O; pyrolysis; terrestrial ecosystem.

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### Introduction

Biochar is created through the pyrolysis of biomass under high-temperature and low-oxygen conditions in which a portion of easily degradable carbon (C) is converted into a more stable or recalcitrant form (Lehmann 2007a, Laird et al. 2009, Spokas and Reicosky 2009). By-products syngas and bio-oil can be utilized as bioenergy with a great potential to substitute fossil fuel. The fixed C content in biochar is recalcitrant to decomposition. A slower returning rate to atmosphere than that in raw biomass was reported (Lehmann et al. 2006, Park et al. 2011, Woolf and Lehmann 2012). This enables long-term storage of carbon, which offsets GHG emissions from fossil fuel. Biochar application can improve soil quality, reduce nutrient losses in run-off, enhance the utilization efficiency of fertilizer, promote agricultural productivity, increase total soil C stocks, and sequester more C from the atmosphere (Ennis et al. 2012). Moreover, biochar can be produced conveniently with various biomass feedstocks in industrial or domestic facilities and thus can be applied globally. Woolf et al. (2010) estimated that global implementation of biochar could potentially offset a maximum of 12% of the current anthropogenic  $CO_2$ -C equivalent ( $CO_2$ -C<sub>e</sub>) emissions. Therefore, biochar application in agricultural soil has been recommended as a strategy to reduce the increasing atmospheric  $CO_2$  concentration and abate climate change (Lehmann 2007*b*, Laird 2008, Woolf et al. 2010, Biederman and Harpole 2013, Liu et al. 2013).

However, there are still uncertainties surrounding the biochar's climate-mitigation potential, particularly the direction and magnitude of reduced soil GHG emissions after biochar application. Three key GHGs that contribute to global warming,  $CO_{2'}$  methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O), continue to increase due to human activities (IPCC 2007). Biochar application can impact soil GHG fluxes by changing the composition and activity of soil microbes, soil pH, and soil biogeochemical processes (Chan et al. 2008, Spokas and Reicosky 2009, van Zwieten et al. 2010*a*). Liu et al. (2011) observed that biochar application decreased CH<sub>4</sub> and CO<sub>2</sub> emissions from waterlogged paddy soil in the laboratory, and they attributed

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such results to the restriction in methanogen activity and limitation on microbial biomass carbon, as well as the rise of pH value. In addition, Rondon et al. (2005) found that biochar amendment reduced N<sub>2</sub>O emissions from pastureland and soybean soil by 80% and 50%, respectively, because microbial conversion and denitrification were restricted. Several studies have reported similar effects of biochar application on soil GHGs emissions (van Zwieten et al. 2010a, Feng et al. 2012). However, the increase in soil CH<sub>4</sub> (Knoblauch et al. 2011, Singla and Inubushi 2014) and CO<sub>2</sub> (Scheer et al. 2011, Ameloot et al. 2013, Felber et al. 2014) emissions were also observed after biochar application. For example, Zhang et al. (2010, 2012a, b) reported that biochar applied at a rate of 40 t-ha-1 decreased N<sub>2</sub>O emission from paddy and maize fields by 21–28% and 10.7-41.8%, respectively, but increased CH<sub>4</sub> emission from a paddy field by 41% and CO<sub>2</sub> emission from a maize field by 12%.

These results show the highly variable effects of biochar amendment on soil GHG fluxes, depending on the study conditions, duration of the experiment, biochar application rate, biochar feedstock, and pyrolysis methods (Mukome et al. 2013). Such variable results substantially weaken the potential of biochar as an option to reduce soil GHG emissions. Cayuela et al. (2014) and Sagrilo et al. (2014) reviewed the effect of biochar application on N<sub>2</sub>O and CO<sub>2</sub> emissions, respectively. Clearly, the three major GHGs should be simultaneously considered when evaluating if a policy is effective to mitigate climate change (Tian et al. 2015). A quantitative and comprehensive literature analyzing the effect of biochar application on the three GHG fluxes together is scarce. The absent knowledge about comprehensive effect on three GHG emissions decreases the predictive accuracy of models calculating reduced soil GHG emissions due to biochar application and limits our understanding of the potential role of biochar in mitigating global climate change (Woolf et al. 2010).

We performed a comprehensive meta-analysis that integrates the results of previous studies that investigated soil GHG fluxes following biochar amendment. The objective of the current study was to test whether the direction and magnitude of the GHG fluxes from soils amended with biochar differed based on the following variables: (1) study type (field experiment vs. laboratory incubation), (2) field type (upland vs. paddy), (3) experiment length (several days to 3 years), (4) biochar application rate (2 to 150 t-ha<sup>-1</sup>), (5) biochar feedstock, and (6) pyrolysis temperature (350 to 900°C) (Cayuela et al. 2014, Sagrilo et al. 2014).

### **Materials and Methods**

#### Data selection

Data were obtained from peer-reviewed publications that reported soil GHG fluxes with or without biochar application. The relevant publications were identified through a keyword search using the terms "biochar," "charcoal," and "char," which were used to query the Web of Science, Google Scholar (scholar.google.com) and the China Knowledge Resource Integrated Databases. Unreplicated studies were excluded from the meta-analysis. The data



**Fig. 1.** Site distribution of studies examining the response of greenhouse gas fluxes and soil organic carbon content to biochar application that were included in the meta-analysis.

were obtained from studies that investigated any one of the three GHGs fluxes in both the control and biocharamended treatments. In this study, the control was defined as being identical to the treatment for all variables but without biochar (Cayuela et al. 2014). To conduct a comprehensive analysis, a total of 61 peer-reviewed publications containing 296 observations from 19 countries across six continents mainly distributed in West Europe, North America, and East Asia (Fig. 1; Appendix S1: Table S1) were selected. The numerical values extracted from figures in selected articles were obtained using an Engauge Digitizer (Free Software Foundation, Boston, Massachusetts, USA). The 296 observations were divided into three categories based on GHG type: CO<sub>2</sub>, N<sub>2</sub>O, and  $CH_4$ . Both emission and uptake of  $CH_4$  were observed. In order to distinguish the actual effects of biochar application, including positive and negative effects on CH emission or uptake, the CH<sub>4</sub> category was further divided into three subcategories based on observations: CH<sub>4</sub> emitted from the biochar treatment and control plots (E-E), CH<sub>4</sub> uptake in the biochar treatment and control plots (U-U), and CH<sub>4</sub> uptake in the control plots but emitted in the biochar-amended treatment plots (U-E). There were no reports on CH<sub>4</sub> emissions from the control and taken by biochar-amended treatment (E-U) plots. In addition, data were collected on the soil type, soil pH, soil organic carbon (SOC) content, and soil total nitrogen (TN) concentration.

To better understand the factors that regulate the direction and magnitude of the GHG responding to biochar application, the observations within each GHG category were subdivided to study type (field experiment vs. laboratory incubation), field type (upland vs. paddy), experiment length ( $\leq 0.5, 0.5-1, 1-2, 2-3$  yr for field experiment;  $\leq 30, 30-60, 60-90, >90$  day for laboratory incubation), biochar application rate ( $\leq 10, 10-20, 20-30, 30-40, >40 \text{ t}\cdot\text{ha}^{-1}$ ), biochar feedstock (wood, straw, husk, poultry manure), and pyrolysis temperature ( $\leq 500, 500-600, 600-700, 700-900^\circ$ C).

### Meta-analysis

The size of the effect for each investigation was calculated as the response ratio r = Xe/Xc, where Xe is the GHG flux in the biochar treatment plots and Xc is the GHG flux in the corresponding control plots. The GHG flux was usually measured frequently or continuously during the experiments, the average or cumulative flux in the publications was transformed into a response ratio.

Because some publications only reported mean values without standard deviations or standard error values, the number of observations in the studies used in this analysis was maximized according to an unweighted meta-analysis as described in previous studies (Guo and Gifford 2002, Knorr et al. 2005, Song et al. 2014). The data were analyzed using METAWIN 2.0 (Rosen-

berg et al. 2000). This procedure is analogous to the partitioning of variance in a standard analysis of variance where total heterogeneity among groups ( $Q_t$ ) was partitioned into within-group ( $Q_w$ ) and between-group ( $Q_b$ ) heterogeneity. The Q statistic follows a chi-square distribution with k - 1 degrees of freedom, with k referring to pairs of means and not separate publications (Hedges and Olkin 1985, Knorr et al. 2005). The  $Q_b$  for each categorical variable was determined for the response variable. A significant  $Q_b$  value indicated that the effect size differed between different categorical subdivisions.

The mean effect size for each categorical subdivision was calculated, and a bias-corrected 95% confidence interval (CI) was determined by applying a bootstrapping procedure using METAWIN software. The effect of biochar application on the soil GHG fluxes within a categorical subdivision was considered significant at P < 0.05 if the 95% CIs did not include 1 (Liu and Greaver 2009). Pearson correlations between the response ratio of the GHG fluxes and soil pH, SOC, and TN contents were determined using SPSS software (version 16.0, SPSS, Chicago, Illinois, USA) with Microsoft Windows.

### Results

### CO<sub>2</sub> flux

The results of CO<sub>2</sub> flux were drawn from 77 observations of 31 publications. In general, biochar application significantly increased CO<sub>2</sub> emission by an average of 19%, with an increase of 5% in the field and 28% in the laboratory (P < 0.05) (Fig. 2a). In the field experiments (Fig. 2B), biochar significantly decreased CO<sub>2</sub> emissions by 5% in paddy fields but increased CO<sub>2</sub> emissions by 12% in upland fields (P < 0.05). CO<sub>2</sub> emissions were not affected over a short time period (<0.5 years) after biochar application but increased by 21% during 0.5 to 1 year. After 1 year, however, this increase slowed down. Low biochar application rates ( $\leq 10 \text{ t} \cdot \text{ha}^{-1}$ ) decreased CO<sub>2</sub> emissions, whereas high application rates (>10 t·ha<sup>-1</sup>) significantly (P < 0.05) increased CO<sub>2</sub> emissions by +11% and +6% at the application rates of 10–20 and 30–40 t·ha<sup>-1</sup>, respectively. Biochar derived from wood (including soft and hard woods) significantly (P < 0.05) increased CO<sub>2</sub> emissions by an average of 21%, whereas biochar derived from husk significantly (P < 0.05) decreased CO<sub>2</sub> emissions. Biochar produced under a low pyrolysis temperature (≤500°C) temperature (700–800°C) high or significantly (P < 0.05) increased CO<sub>2</sub> emissions, whereas biochar produced at temperatures ranging from 500 to 600°C had no significant effect on CO<sub>2</sub> emission (P > 0.05).

In laboratory incubations (Fig. 2c), the most positive priming effect on  $CO_2$  emissions occurred



**Fig. 2.** Untransformed response ratios pertaining to the effects of biochar application on  $CO_2$  emissions based all data (a), field (b), and laboratory (c) experiments. Dots with error bars denote the overall mean response ratio and the 95% Cls. Different capital letters to the right of the bars indicate statistically significant differences at the *P* < 0.05 level.

during the initial 60 days after biochar application (P < 0.05). Moreover, this positive effect generally became stronger with higher biochar application rates. Only the biochar produced from husk significantly (P < 0.05) decreased CO<sub>2</sub> emissions, whereas the other three biochar types significantly increased CO<sub>2</sub> emissions. Biochar produced at low pyrolysis temperatures ( $\leq 600^{\circ}$ C) significantly (P < 0.05) increased CO<sub>2</sub> emissions, whereas biochar produced at high temperatures ( $\geq 600^{\circ}$ C) significantly (P < 0.05) decreased CO<sub>2</sub> emission.

### N<sub>2</sub>O flux

The results were drawn from 177 observations of 51 publications. Most of these observations were from laboratory incubations, with only 31% of data obtained

from field experiments. Biochar application significantly (P < 0.05) decreased N<sub>2</sub>O emissions, by 19% and 15%, in the field and laboratory experiments, respectively, with an average of 16% (Fig. 3a). In both upland and paddy fields (Fig. 3b), biochar amendment significantly decreased N<sub>2</sub>O emissions (P < 0.05). More than 76% of the field experiments testing the effect of biochar on N<sub>2</sub>O emission were conducted for less than 0.5 years and in which the N<sub>2</sub>O emissions were observed to significantly (P < 0.05) decrease by 21%. Only one field experiment was performed for more than 2 years, where biochar stimulated N<sub>2</sub>O emissions. The suppressive effect of biochar on N<sub>2</sub>O emissions generally increased with the biochar application rate. All biochar types significantly (P < 0.05) decreased N<sub>2</sub>O emissions. Biochar produced under a low pyrolysis temperature  $(\leq 600^{\circ}\text{C})$  significantly (P < 0.05) decreased N<sub>2</sub>O



**Fig. 3.** Untransformed response ratios pertaining to the effects of biochar application on N<sub>2</sub>O emissions based on all data (a), field (b), and laboratory (c) experiments. Dots with error bars denote the overall mean response ratio and the 95% CIs. Different capital letters to the right of the bars indicate statistically significant differences at the P < 0.05 level.

emissions, whereas high pyrolysis temperature (700–900°C) did not significantly influence N<sub>2</sub>O emissions.

In laboratory incubations (Fig. 3c),  $N_2O$  emissions significantly (P < 0.05) decreased during the initial 30 days or after 90 days following biochar application, whereas no significant difference was observed during the 30- to 90-day period. Biochar application rates, low ( $\leq 10 \text{ t-ha}^{-1}$ ) or high (>40 t-ha^{-1}), significantly (P < 0.05) reduced  $N_2O$  emissions. Only wood biochar treatments significantly (P < 0.05) decreased  $N_2O$  emission. Application of biochars derived from low pyrolysis temperatures ( $\leq 500^{\circ}$ C) significantly (P < 0.05) reduced soil  $N_2O$  emission.

#### CH₄ flux

The results were drawn from 42 observations of 19 publications. In the E-E group including 31

observations (Fig. 4a), biochar application significantly (P < 0.05) increased CH<sub>4</sub> emission by 19% under field conditions, but significantly (P < 0.05)decreased CH<sub>4</sub> emission by 18% in laboratory incubations. In the E-E field experiments (Fig. 4b), all 24 observations were obtained from paddy fields. A slight suppressive effect of biochar on CH<sub>4</sub> emissions was observed during the initial 60–120 days after biochar application, but a significant (P < 0.05) increase (+41%) occurred during the 120- to 180-day period. A biochar application rate of 20-30 t-ha-1 significantly (P < 0.05) decreased CH<sub>4</sub> emission by 50%, whereas other application rates significantly (P < 0.05) increased CH<sub>4</sub> emissions, especially the rate of 10-20 t·ha<sup>-1</sup> (+79%). Biochar derived from straw and husk significantly (P < 0.05) increased CH<sub>4</sub> emission. Biochar significantly (P < 0.05) increased CH<sub>4</sub> emission by 39% at pyrolysis temperatures



**Fig. 4.** Untransformed response ratios pertaining to the effects of biochar application on  $CH_4$  emissions based on all E-E data (a), E-E field data (b), U-U data (c), and U-E data (d). E-E: $CH_4$  was emitted from the biochar-treated and control plots; U-U: $CH_4$  was consistently taken up in the biochar-treated and control plots; U-E: $CH_4$  was taken up in the control plots but emitted from the biochar-treated plots. Dots with error bars denote the overall mean response ratio and the 95% CIs. Different capital letters to the right of the bars indicate statistically significant differences at the P < 0.05 level.

ranging from 400 to 500°C, but significantly (P < 0.05) decreased by 31% at lower (300–400°C) pyrolysis temperatures.

All of the upland field experiments in the literature exhibited  $CH_4$  uptake in the control plots. After biochar treatment, however,  $CH_4$  fluxes varied. In the U-U group (Fig. 4c), which comprised a total of six observations from four upland fields and two laboratory incubations, biochar application significantly (P < 0.05) enhanced  $CH_4$  uptake by 114% under field conditions but decreased  $CH_4$  uptake by 24% under laboratory incubations. In the U-E group (Fig. 4d), all of three upland field experiments showed net  $CH_4$  uptake in the control soil but converted into net  $CH_4$  emission after biochar amendment. Due to limited observations, data from the U-U and U-E experiments and the E-E laboratory incubations were not analyzed further.

# Factors controlling GHG responses to biochar application

The statistical analysis of between-group heterogeneity  $(Q_b)$  showed that the response of CO<sub>2</sub> emissions to biochar application differed significantly (P < 0.05) between the field and laboratory studies, in contrast to N<sub>2</sub>O and CH<sub>4</sub> emissions (Table 1). In the field experiments, field type (upland/paddy) significantly (P < 0.05) affected the response of CO<sub>2</sub> emissions to biochar application, and the duration of the experiment significantly (P < 0.05) affected CH<sub>4</sub> emissions. The biochar application rate significantly (P < 0.05) influenced CH<sub>4</sub> emissions, and biochar feedstock significantly (P < 0.05) impacted CO<sub>2</sub> and N<sub>2</sub>O emissions. In addition, the pyrolysis temperature of the biochar significantly (P < 0.01) affected CH<sub>4</sub> emissions.

GHGs	Study type	Categorical variable	$Q_{b}$	Р	F
CO <sub>2</sub>	Total	Field/laboratory	0.922 0.014		6.358
	Field	Upland/paddy	0.199	0.026	5.489
		Experiment length	0.099	0.517	0.778
		Biochar addition rate	0.166	0.429	0.993
		Biochar feedstock	0.356	0.026	3.589
		Pyrolysis temperature	0.194	0.091	2.617
	Laboratory	Experiment length	1.012	0.194	1.644
		Biochar addition rate	2.513	0.013	3.620
		Biochar feedstock	2.892	0.002	6.011
		Pyrolysis temperature	3.566	0.0002	8.236
N <sub>2</sub> O	Total	Field/laboratory	0.062 0.727		0.122
	Field	Upland/paddy	0.003	0.892	0.019
		Experiment length	0.146	0.828	0.297
		Biochar addition rate	0.598	0.446	0.944
		Biochar feedstock	1.276	0.039	2.995
		Pyrolysis temperature	0.668	0.086	2.582
	Laboratory	Experiment length	7.357	0.010	3.946
		Biochar addition rate	1.48	0.771	0.451
		Biochar feedstock	6.030	0.031	3.077
		Pyrolysis temperature	8.134	0.007	4.265
$CH_4$	E-E	Field/laboratory 0.741		0.117	2.617
	Field	Upland/paddy	—	—	_
		Experiment length	1.109	0.030	5.369
		Biochar addition rate	3.220	0.002	6.282
		Biochar feedstock	0.250	0.324	1.019
		Pyrolysis temperature	1.688	0.026	4.419
	U-U	Field/laboratory	2.521	0.346	1.139
	U-E	Field/laboratory	13.831	0.325	1.381

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Notes: E-E:CH<sub>4</sub> was emitted from the biochar-treated and control plots; U-U:CH<sub>4</sub> was consistently taken up; U-E:CH<sub>4</sub> was taken up in the control plots and emitted from biochar treatment plots. Bold values is to highlight the P value (<0.05).

In the laboratory experiments, the biochar application rate, biochar feedstock, and pyrolysis temperature significantly (P < 0.05) affected CO<sub>2</sub> emissions. N<sub>2</sub>O emissions were significantly (P < 0.05) influenced by the duration of the experiment, biochar feedstock, and pyrolysis temperature.

Pearson correlation analysis (Appendix S1: Table S2) showed that the response ratio of CH<sub>4</sub> emissions had a positive correlation with the SOC content (P < 0.01), a negative correlation with soil pH (P < 0.01), and no significant correlation with TN content of the soil in the E-E field experiment. No significant correlations were observed between response ratios of CO<sub>2</sub> or N<sub>2</sub>O emissions and the SOC content, soil pH, or the TN concentration of the soil in field experiments or laboratory incubations (P > 0.05). The relationships between the response

ratios of the GHG fluxes after biochar application and climatic factors such as the mean annual temperature and precipitation of the study sites were not analyzed because these data were not provided in most of the selected publications.

## Discussion

### Effect of biochar application on soil CO<sub>2</sub> emissions

Soil CO<sub>2</sub> fluxes are produced mainly by soil microorganism and plant root respiration (Hanson et al. 2000). Our analysis showed that biochar amendments significantly stimulated CO<sub>2</sub> emissions from upland fields and laboratory incubations but reduced CO<sub>2</sub> emissions from paddy fields. One of possible reasons is that

biochar amendments may increase the biomass and activities of microorganisms and thus enhance the decomposition of native soil organic matter (SOM) (Wardle et al. 2008, Steinbeiss et al. 2009). Another possible reason is that a portion of the labile organic carbon pool of amended biochar may have been consumed by microorganisms, resulting in increased CO<sub>2</sub> emissions (Luo et al. 2011). In addition, biochar application increases plant growth and root biomass (Major et al. 2010, Lehmann et al. 2011), which may promote root respiration and provide additional organic matter for decomposition. These factors increase soil CO<sub>2</sub> emissions with the rate and time of biochar application. However, due to anaerobic conditions in paddy fields, biochar application may stimulate CH<sub>4</sub> production (Knoblauch et al. 2011) and consequently reduce CO, emissions. An increase in CO<sub>2</sub> emissions may depend on the organic C content of the soil (Sheng et al. 2010). For example, Kimetu and Lehmann (2010) observed that biochar amendment increased CO<sub>2</sub> emissions from SOC-rich soil but reduced CO<sub>2</sub> emissions from SOCpoor soil. However, the current comprehensive analysis did not detect a significant correlation between the response ratio of CO<sub>2</sub> emissions to biochar amendment and SOC content (P > 0.05).

Because biochar is a typical alkaline substance, biochar amendments usually increase soil pH and subsequently increase the solubility of  $CO_2$  and the formation of bicarbonate acid (Jensen 2003), leading to a reduction in  $CO_2$  emissions, especially in paddy fields. However, the liming effects of biochar tend to disappear with time, resulting in a decrease in soil pH (Slavich et al. 2013, Cayuela et al. 2014) and subsequent release of  $CO_2$  from carbonic acid. The combination of this released  $CO_2$  and the  $CO_2$  emitted from microbial respiration and root respiration results in a peak  $CO_2$  flux at a certain time period following biochar amendment, i.e., from 0.5 to 1 year in this analysis (Fig. 2b).

# Effect of biochar application on soil N<sub>2</sub>O emissions

Soil  $N_2O$  emissions result from three main processes: nitrification, denitrification, and dissimilatory nitrate reduction (Baggs 2011). Agricultural soil is the main source of global anthropogenic  $N_2O$  emissions due to the widespread use of synthetic nitrogen (N) fertilizers, and denitrification contributed the most  $N_2O$  release (Dalal et al. 2003). Our analysis showed that biochar application significantly suppressed  $N_2O$  emissions, especially in field experiments.

In a model calculating global annual avoided soil  $N_2O$  emissions from biochar-amended cropland, Woolf et al. (2010) assumed the reduction factor ( $R_N$ ) to be 25%. A recent meta-analysis reported that biochar reduced soil  $N_2O$  emissions by 28% in the field (Cayuela et al. 2015). Our current study showed that soil  $N_2O$  emission was

generally reduced by 19% after biochar application in the field experiments.

There are several possible mechanisms to explain this result. Firstly, biochar contains considerable amounts of soluble base cations that can neutralize soil acidity (Yuan et al. 2011) increasing soil pH, and shift the product stoichiometry of denitrification resulting in an increased production of N<sub>2</sub> relative to N<sub>2</sub>O (van Zwieten et al. 2010b, Cayuela et al. 2013), and facilitate the reduction of greater amounts of N<sub>2</sub>O to N<sub>2</sub> (Yanai et al. 2007). The acid-neutralizing (liming) capacity of biochar varies with feedstock type and pyrolysis temperature (Cao et al. 2009, Singh et al. 2010, Yuan et al. 2011). This variability in biochar properties can explain the variation in N<sub>2</sub>O emissions following the application of biochar. The liming effect of biochar becomes weaker and the soil pH in the vicinity of biochar particles decreases with time (Slavich et al. 2013, Cayuela et al. 2014). This effect was also supported by the results of our analysis that showed that the significant suppression of N<sub>2</sub>O emissions only occurred during the early stages of biochar application and tended to weaken overtime (Fig. 3).

Secondly, biochar amendments increase soil porosity and aeration by absorbing excess soil moisture (Yanai et al. 2007) and reducing soil compaction and bulk density (Rogovska et al. 2011), which decreases  $N_2O$  generation (Heincke and Kaupenjohann 1999, Richardson et al. 2009, Case et al. 2012).

Thirdly, biochar may restrict N availability, a major driver of soil N<sub>2</sub>O release, and therefore suppress N<sub>2</sub>O generation (Cayuela et al. 2014). The biochar surface can sorb  $NO_2^-$  and thus decrease  $N_2O$  emissions (Cheng et al. 2008, Cayuela et al. 2013, Clough et al. 2013). Mukherjee and Zimmerman (2013) demonstrated that biochar produced at higher pyrolysis temperatures had greater sorption due to a greater surface area compared with biochar produced at lower temperatures. However, this effect does not explain our results of increased N<sub>2</sub>O emissions in response to application of biochar produced at relatively high temperatures (Fig. 3), which implied the complexity of pyrolysis temperature effect on N<sub>2</sub>O emissions. Nitrate sorption increased with biochar application rate (Cayuela et al. 2013) but decreased over time due to oxidation (Cheng et al. 2008, Lin et al. 2012). This finding is consistent with our results showing that N<sub>2</sub>O emissions gradually decreased with higher biochar application rates, especially in field studies, and this suppression of N<sub>2</sub>O emissions became weaker over time.

The biochar was investigated to determine the presence of potential inhibitory or toxic compounds that can restrain microbial communities. Such compounds are released within a short-term period after biochar application to soil (Spokas et al. 2010, Cayuela et al. 2014). This potential mechanism, which requires further verification, may also explain the initial suppression of  $N_2O$  emissions after biochar application in the current study.

The suppressive effect of biochar application on N<sub>2</sub>O emissions can influence the soil N cycle through several possible mechanisms as noted above. DeLuca et al. (2006) found that biochar can readily enhance nitrification, likely through an alteration of the activity of the nitrifying community or via the elimination of inhibitory compounds, or via liming (Wang et al. 2014), rather than a simple reduction in NO<sub>3</sub><sup>-</sup> immobilization potential. Adsorption of various forms of N, such as  $NO_3^-$  and  $NH_4^+$ on biochar could improve N utilization efficiency, increase crop yield, and reduce N losses from agricultural soil to waterbody and atmosphere (Pan et al. 2013). However, Biederman and Harpole (2013) found that biochar application did not affect soil available N but increased total soil N. Therefore, studies focusing on the N cycle after biochar application are worth further investigation.

### Effect of biochar application on soil CH<sub>4</sub> emissions

During methane production, organic matter is used as a substrate under anaerobic conditions and is consumed by methanotrophic bacteria before being released to the atmosphere under aerobic conditions (Le Mer and Roger 2001, Watanabe et al. 2007).  $CH_4$  emissions result from a balance between  $CH_4$  production and oxidation. Biochar amendments may improve soil aeration, and thus decrease  $CH_4$  production and/or increase  $CH_4$  oxidation (van Zwieten et al. 2009), which explains the greater net uptake of  $CH_4$  in the presence of biochar in the four upland field experiments in the current study (Fig. 4C).

In the paddy fields, the aeration effect of biochar may be temporary and may disappear over time because of water logging. Consequently, the suppression of CH<sub>4</sub> emissions disappears. However, biochar application may also increase microbial biomass and activity (O'Neill et al. 2009, Steinbeiss et al. 2009), which would enhance the decomposition of native SOM (Wardle et al. 2008) in addition to the decomposition of the labile organic C pool of biochar. These decomposition processes provide the predominant substrates for methanogens and stimulate the growth of methanogenic archaea (Feng et al. 2012). In turn, this activity promotes CH<sub>4</sub> production (Knoblauch et al. 2011) and a consequent decrease in CO<sub>2</sub> emissions. These processes provide a likely explanation for the results of our analysis, which revealed a reduction in CH<sub>4</sub> emissions shortly after biochar application, followed by a significant increase.

Biochar can efficiently retain ammonium  $(NH_4^+)$  through adsorption (Liang et al. 2006). Ammonium and  $CH_4$  compete for oxidation by methanotrophs, and therefore, the presence of  $NH_4^+$  can stimulate  $CH_4$  emissions from paddy fields (Mosier et al. 1991). This sorption increases with the biochar application rate, which is consistent with our results showing that  $CH_4$  emissions generally increased with the biochar application rate in paddy field studies.

However, our conclusions are based on short-term (<6 months) observations in paddy fields and a limited number of observations in upland fields. Consequently, the long-term effects of biochar application on  $CH_4$  fluxes remain unclear and urgently require further long-term investigation.

# Factors affecting greenhouse gas emissions from biochar-amended soils and their uncertainty

The effects of biochar application on GHG fluxes varied considerably in response to multiple factors, resulting in different and even contrasting results. The observations on positive effect on CO<sub>2</sub> emission was overestimated but suppressive effect on N<sub>2</sub>O emission was underestimated in laboratory incubations compared with field experiments. This discrepancy can be attributed partially to experimental conditions and the duration of the experiments. Contradictory responses of CH<sub>4</sub> fluxes to biochar application were obtained from the field experiments and laboratory incubations. In the published literature, most of the effects of biochar amendments on GHG fluxes, especially N<sub>2</sub>O fluxes, were obtained from laboratory incubations (Appendix S1: Table S1) rather than field experiments. Extrapolating conclusions drawn from short-term laboratory incubations is unwarranted because field experiments can provide more reliable and practical results for recommending field-scale biochar application to mitigate climate change. In addition, the water content of the soil also plays a key role in determining the effect of biochar application. For instance, the responses of CO<sub>2</sub> and CH<sub>4</sub> emissions to biochar application were opposite between upland and paddy fields. The difference in water-filled pore space (WFPS) or water holding capacity (WHC) in laboratory incubations resulted in varying effects of biochar on GHG fluxes (Appendix S1: Table S1). Cayuela et al. (2014) also found that soil moisture influenced biochar N<sub>2</sub>O mitigation capacity.

The responses of GHG fluxes to biochar amendments vary considerably over time. Most studies on GHG fluxes were conducted for less than 6 months in field experiments and less than 90 days in laboratory incubations (Appendix S1: Table S1). The reliability of these results needs to be further verified through long-term investigations.

The responses of GHG fluxes varied significantly with the biochar application rate. The biochar added at a rate of 20–30 t·ha<sup>-1</sup> had the strongest suppressive effect on emissions of the three GHGs in field experiments. To mitigate GHG emissions, this application rate is feasible and can be recommended for future application.

In addition, biochar feedstock had a significant effect on the GHG fluxes. In most studies, biochar produced from wood or straw was applied. Other types of biochar, i.e., husk and poultry manure, were used in some

	Change (kg·ha⁻¹)			GWP (N O+CH )	CO -C equivalent (CO -C )
Field type	CO <sub>2</sub>	N <sub>2</sub> O	CH <sub>4</sub>	$(kg\cdot ha^{-1})$	$(kg \cdot ha^{-1})$
Upland	+2017.48 ± 592.13	$-2.21 \pm 0.62$		-658.58	+370.61
Paddy	$-670.37 \pm 124.08$	$-2.35 \pm 0.66$	51.96 ± 19.17	+598.7	-19.55

**Table 2.** The change (mean ± SE) of annual emissions of CO<sub>2</sub>, N<sub>2</sub>O, and CH<sub>4</sub> from the field and global warming potential (GWP) after biochar amendment.

*Notes:* "+"mean the increase in emissions, "-" mean the decrease in emissions. The IPCC GWP factors (mass basis) for N<sub>2</sub>O and CH<sub>4</sub> are 298 and 25 times higher than CO<sub>2</sub> over the 100-year time horizon, respectively (IPCC 2007).

experiments, especially laboratory incubations. Our analysis showed that biochar produced from husk had the strongest suppressive effect on  $CO_2$  and  $N_2O$  emissions (Figs. 2, 3). Therefore, husk may be considered as a preferred feedstock for biochar production.

Biochar stability and the interaction between biochar and soil biota vary with pyrolysis temperature (Lehmann et al. 2011, Wang et al. 2015). In the field experiments, application of biochar produced at pyrolysis temperatures between 500 and 600°C had no significant effect on  $CO_2$  emission. Significantly (P < 0.05) increased CO<sub>2</sub> emissions were observed from soils applied with biochar produced either at lower pyrolysis temperatures (≤500°C) or higher temperature (700– 800°C). However, biochar produced at pyrolysis temperatures less than 500°C or 400°C suppressed N<sub>2</sub>O and  $CH_4$  emissions, respectively. It has been reported that increase of pyrolysis temperature from 200°C to 700°C led to consistent decreases in the molar H:C ratio of biochar (Klüpfel et al. 2014), which resulted in a greater reduction in N<sub>2</sub>O emissions (Cayuela et al. 2015). Biochar pyrolyzed at low temperatures has a higher H:C ratio and is more rapidly degraded by soil microorganisms (van Zwieten et al. 2010a), and this may facilitate the mineralization of organic matter (Luo et al. 2011, Zimmerman et al. 2011) and induce more CO<sub>2</sub> emission. Biochar pyrolyzed at higher temperatures can form more condensed aromatic structures that are resistant to microbial decomposition (Glaser et al. 2002, Liang et al. 2006). Wang et al. (2015) reported that biochars produced at the low pyrolysis temperatures (200–375°C) induced significant (P < 0.05) priming effect on SOM mineralization, whereas biochars produced at higher pyrolysis temperatures (>400°C) did not significantly impact SOM mineralization. These results indicated that the effect of pyrolysis temperature on SOM mineralization can only limitedly account for soil CO<sub>2</sub> emission.

There were only 42 observations for  $CH_4$  fluxes in the current study. The limited data make it difficult to precisely estimate the potential tradeoff among the three GHG fluxes following biochar application. The uncertainty of the results was further increased by the difference of duration of the experiments (<6 months in most studies), study type (fewer field experiments vs. more laboratory incubations), and pyrolysis temperature data (mostly less than 500°C). Some other factors, such as biochar chemical properties, soil type and pH, crop strains, fertilizer application, and tillage methods, can also impact the effects of biochar application on soil GHG emissions (Biederman and Harpole 2013, Cayue-la et al. 2014). For example, N fertilizer application is usually a critical factor controlling  $N_2O$  emission, which even has stronger effect on  $N_2O$  emission than biochar application. Due to the limited data in the selected publications, these analyses were not been performed in the present study.

Application of any biomass to soil will likely change GHG fluxes. A better comparison should be based on the effects of an equivalent amount of biomass that was applied into the soil either directly or after conversion into biochar (Schimmelpfennig et al. 2014, Cayuela et al. 2015), which reflect the actual effects of biochar application to agricultural soil and provide true reference for farmers and policy makers as an alternative choice. Unfortunately, most of literatures did not provide the data about the effect of biomass added into soil directly, which resulted in the inherent bias of the present analysis.

The global warming potential (GWP) of CH<sub>4</sub> and N<sub>2</sub>O is 25 and 298 times higher than CO<sub>2</sub> over the 100-year time horizon, respectively (IPCC 2007). Based on the result of the meta-analysis, the change in annual soil emissions of  $CO_2$ ,  $N_2O_2$ , and  $CH_4$  and GWP after biochar amendment was estimated (Table 2). CH<sub>4</sub> flux from upland was not considered because of the small quantity and high uncertainty. In general, CO<sub>2</sub>-C equivalent (CO<sub>2</sub>-C<sub>2</sub>) emissions would increase 370.61 kg·ha<sup>-1</sup>·yr<sup>-1</sup> if biochar was applied to upland field, but would reduce 19.55 kg·ha<sup>-1</sup>·yr<sup>-1</sup> if biochar was applied to paddy field. The global total area of upland and paddy field was  $12 \times 10^8$  ha and  $1.5 \times 10^8$  ha, respectively (Liu and Greaver 2009). Annual CO<sub>2</sub>-C<sub>e</sub> emissions would increase 444.73 Gg or reduce 2.93 Gg when biochar was applied to all upland field or paddy, respectively. We may recommend biochar application to paddy fields as an approach to mitigating climate change. In upland agricultural fields that cover larger areas, however, biochar application should be considered with caution. It is important to note that these suggestions are based only on the effects on soil GHG fluxes. The potential role of biochar in mitigating climate change

still requires a comprehensive estimation combined with slowed biomass decomposition, improving crop productivity, and soil carbon storage (Woolf et al. 2010, Jeffery et al. 2011, Biederman and Harpole 2013, Liu et al. 2013). Moreover, most of observation was conducted in one growing season (about 3 to 5 months) (Appendix S1: Table S1), which will bring considerable risk when applying the short-term results to the annual scale.

## Conclusions

The effects (stimulation or suppression) of biochar amendment on soil GHG fluxes varied with the characteristics and application rates of biochar, and the soil conditions. Biochar amendments significantly reduced N<sub>2</sub>O emissions both in the field and the laboratory experiments, significantly decreased CO, emissions from paddy field but increased CO<sub>2</sub> emissions from both upland fields and laboratory incubations. Biochar application significantly increased CH<sub>4</sub> emissions from paddy fields but reduced CH<sub>4</sub> emissions in laboratory incubations. After considering the tradeoff among the three GHG fluxes, biochar amendments largely reduced CO<sub>2</sub>-C equivalent emissions from paddy fields but increased CO<sub>2</sub>-C equivalent emissions from upland fields. Only taking account of effects on soil GHG fluxes, biochar application to paddy fields can be confidently recommended as a priority to mitigate climate change, whereas biochar application to upland fields should be considered with caution. However, these effects highly varied over time and were influenced by the biochar application rate, feedstock, and pyrolysis temperature. Moreover, the longterm effects of biochar amendments on GHG fluxes remain unclear, which challenges our previous understanding obtained mainly from short-term laboratory incubation. It should be prudent to extrapolate the findings from short-term experiments. Therefore, the potential of biochar application to cropland to mitigate climate change should be further investigated in more long-term field experiments across regions and biomes.

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