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Impact of land use and soil properties on soil methane flux response to biochar addition

Weiwei Cong [1], Jun Meng [1]*, Samantha C. Ying[2]*

[1] *College of Agronomy, Shenyang Agricultural University, Shenyang, 110161, China*

[2] *Department of Environmental Science, University of California Riverside, Riverside, CA 92507, USA*

* Corresponding author.

E-mail address: Jun Meng (mengjun1217@163.com), Samantha C. Ying (samantha.ying@ucr.edu).

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Abstract

Addition of biochar to soils has been shown to increase crop yield and aid in mitigating greenhouse gas emissions by decreasing the extent of soil methane (CH_4) flux. Previous studies utilizing meta-analysis to better understand the impact of environmental and management factors on CH_4 flux from biochar treated soil systems have provided contrasting results, ranging from significant increase, decrease, to no change in methane flux after amendment. We hypothesized that these discrepancies could be explained by separating studies into two major land use categories, upland and paddy, prior to analysis so that the overall redox conditions are more comparable across studies upon which statistical comparisons are made. Furthermore, past studies did not consider potentially critical soil properties including soil organic carbon, total nitrogen, C/N, and soil texture; a number of biochar properties including biochar pH and C/N; and five additional management and experimental factors. In this study, Hedge's *d* metric was calculated and Wilcoxon analyses were used in a meta-analysis to determine the impact of these additional factors on methane flux from biochar-amended upland versus paddy soils. We demonstrate that variations in soil characteristics including SOC, C/N, and pH significantly influences the methane flux from biochar treated soils, while biochar characteristics and management practices have less to no effect as determined by the magnitude of the Hedge's *d* metric. Soils with low SOC, total nitrogen, C/N, acidic or alkaline pH exhibited lowest CH_4 emission rates/highest CH_4 uptake rates, whereas soils with higher SOC content, C/N, and circumneutral pH exhibited higher CH_4 emission with biochar addition. Several possible mechanisms are suggested to explain the role of these variables in CH_4 cycling. Results from this study will be used to evaluate the input parameters for building a linear additive model to quantitatively predict soil methane flux in response to biochar additions. Ultimately, implementation of the linear additive model can be extremely valuable for advising agricultural practices toward minimize methane emissions or maximizing methane sink strength. We suggest that additional field and controlled experiments be performed to better define the reaction network that controls methane flux from biochar treated soils, with particular attention to paddy soils where studies are still lacking.

1 Introduction

Biochar is commonly used as an agricultural amendment to increase crop yield and has also been applied to soils to decrease greenhouse gas emissions (Lehmann et al., 2011). Biochar is produced under low oxygen or anoxic conditions and is highly resistant to decomposition, making its residence time much greater than other plant-derived soil inputs (Sohi et al., 2010; Keiluweit et al., 2010). Because of biochar's recalcitrance, it has the capacity to alter soil redox conditions and microbial activities over long periods, ultimately influencing the rate of greenhouse gas emissions from soils (Woolf et al., 2010).

Methane is a potent greenhouse gas that contributes approximately 30% of the total net anthropogenic radiative forcing of 1.6 W m^{-2} (Solomon et al., 2007). Atmospheric CH_4 increased at a rate of 12 ppb $year^{-1}$ in the 1980s, and has continued to increase significantly since 2008 (Conrad, 2009; Rigby et al., 2008). It has been estimated that addition of biochar to soils may decrease methane emissions by up to 34 Tg per year (Woolf et al., 2010). However, because soil processes altered by biochar addition are poorly defined and its benefits are unclear, farmers and land companies may be reluctant to adopt the use of biochar (Gwenzi et al., 2015). Further elucidation of the soil mechanisms affected by biochar addition is needed to guide its use in agricultural applications.

Numerous controlled experiments have been conducted to examine the response of soils to biochar addition; the results of many of these studies have been summarized in several recent reviews (He et al., 2016;

Jeffery et al., 2016; Song et al., 2016). As revealed by the results from these and other reports, methane response to biochar addition can vary significantly, ranging from total inhibition of methane emission to stimulation of methane production (Scheer et al., 2011; Zhang et al., 2010). To examine the contribution of soil, biochar, and management factors in explaining variable response of soils to biochar addition, Jeffery et al. (2016) performed a meta-analysis of existing experimental results using the standardized mean metric, Hedge's *d*, to compare the impact of various soil, biochar, and management factors on CH_4 flux across 193 studies. The benefit of applying Hedge's *d* over other metrics is the ability to take negative flux values into consideration (i.e., where addition of biochar leads to a methane sink or decrease in methane flux). Jeffery et al. (2016) showed that a number of factors including soil water regime (flooded, non-flooded, wet-dry cycled) and soil pH have significant influence on the impact of biochar additions on methane flux as compared to controls without biochar addition. However, these findings are inconsistent with findings by Song et al. (2016), which concluded that biochar additions to paddy (i.e., flooded) soils could cause up to 19% greater CH_4 emissions than unamended controls. Jeffery et al. noted that the discrepancy in results between the meta-analyses could be due to the use of different analytical metrics. However, we hypothesized that these contrasting results may also be explained by examining the effect of specific soil and biochar properties and management practices on flood (paddy) and non-flooded (upland) farmlands, separately. In this way, the overall redox condition within the soils of each pairwise comparison (observations before and after biochar addition) is more comparable. By applying quantitative meta-analysis based upon land use, we are able to examine the role of additional soil properties (organic C, total N, C:N ratio), biochar properties, (organic C content, pH and pyrolysis temperature), and management factors (biochar and fertilizer application rate) in paddy versus upland soils. Additionally, we use locally-weighted linear regression to examine the trends in methane flux as a function of each variable, the results of which can be used to build linear additive models to predict soil methane flux in response to biochar additions.

2 Methodology

2.1 Data sources

A literature search was conducted using Scopus, Web of Science and Google Scholar databases using the keywords "biochar" OR "charcoal" OR "black carbon" AND "CH_4" OR "methane" OR "greenhouse gas" taking all publications published before July 2016. A total of 268 experimental treatments from 50 peer-reviewed articles published between 2009 and 2016 were selected, inclusive of pot and incubation experiments and field-based studies. For each article the title and abstract were evaluated to verify if they reported original quantitative data on CH_4 emissions and these articles were examined in detail for quality criteria. A minimum of three replicates per treatment was required for the study to be included in the meta-analysis. Studies where gas sampling frequency was considered not appropriate (i.e., gas samples were taken only once or twice during the entire experiment) were not included. Data was collected on studies that compared CH_4 emissions/uptake between a control and a biochar treatment, where the control was defined as being identical to the treatment for all variables except biochar addition.

From each study data were extracted for (i) soil properties (texture, pH, soil organic carbon (SOC), and total nitrogen (TN), (ii) biochar properties (feedstock, production temperature, pH, and C/N ratio), and (iii) management practices and study design (field/pot/incubation study; biochar application rate; study duration; N, P_2O_5 and K_2O-fertilizer application rate). Plot Digitizer 2.6.6 was used to extract data points that were only provided in figures. When necessary, we contacted authors for information on parameters that were missing in the publications; if we were unable to attain the missing data, the study was excluded from the data analysis. If data from the same experiment and study period were reported in several papers (e.g., in chronosequence studies with different papers utilizing data from the same experiment) only data from the longest study was included.

2.2 Data standardization

Data were subjected to a standardization process to allow for comparisons across studies. To examine the effect of land use as a major control on methane flux from biochar amended soils, compiled data were grouped as "paddy soil" or "upland" for the meta-analysis. The criteria for inclusion in these categories are as follows: (i) "paddy soil" is defined as land used for cultivating rice continuously, while (ii) "upland soil" are soils that are not continuously flooded for extended periods of time, including forest, grassland, wildland, and farmland except rice paddies. After separating studies into the two major land use categories, data was compiled on soil and biochar properties and management practices within each study. Each variable was separated into interval or nominal categories, where intervals were determined based on data distributions. The data distribution of each variable is provided in supplementary information and category definitions are as follows. Soil texture was grouped into three categories: (i) coarse (sandy loam, sandy clay loam, loamy sand), (ii) medium (clay loam, loam, silty clay loam, silt, silt loam) or (iii) fine (clay, silt clay, sandy clay) (USDA, 1999). Soil pH values measured with $CaCl_2$ were transformed to be able to compare pH values acquired using distilled water using Equation *(1)* (Biederman and Harpole, 2013):

$$pH[H_2O] = 1.65 + 0.86 \times pH[CaCl_2] \qquad (1)$$

Soil pH, SOC, TN and C/N data were then separated into a number of categories defined by data distribution (Fig. S1-S3).

A similar data processing procedure was performed on biochar properties where values were grouped into categories based on data distribution. Biochar pyrolysis temperatures were grouped into three temperature ranges (≤400, 401-500, 501-600, >600°C). When temperature was reported as a range in the original study (e.g., 500-600°C), the average value was chosen (i.e. 550°C). Feedstocks were grouped into five categories: (i) biosolids (sewage sludge from water treatment plants), (ii) manures or manure-based materials (poultry, pig or cattle), (iii) wood (oak, pine, willow, sycamore and unidentified wood mixtures), (iv) herbaceous plant materials (green waste, bamboo, straws), and (v) lignocellulosic waste (rice husk, nuts shells, paper mill waste). Biochar pH ranged from 6.2 to 10.5 in soils, being predominantly alkaline, and were grouped into four categories (<7, 7.0-8.0, 8.0-10.0, >10). Biochar TOC, TN and C/N (<50, 50-100, >100) were also grouped based on data distribution (Fig. S4-S6).

Biochar application rates were transformed into percentage of dry weight ratio (w:w soil) where the weight of soil was calculated using the height of the soil layer in which biochar was added (or a height of 15 cm when no value is reported) and the bulk density (BD) of the soil. If BD was not provided, it was calculated from the soil texture according to Saxton et al. (1986). Biochar application rate was then grouped into five categories (<1, 1-<2 , 2–<5, 5–<10, ≥10%, dry weight ratio (w:w) basis). Experimental method was grouped into three categories (Field, Pot and Incubation). Finally experimental time (<60, 60-120, 121-240, >240), N, P_2O_5 and K_2O fertilizer were grouped based on data distribution (Fig. S7-S9): no application, <150, >=150 kg ha^{-1}.

2.3 Data analysis

Hedge’s *d* was used as a metric to compare between studies to determine the change in soil methane flux after biochar amendment in various studies. Use of this standardized mean difference metric has been successfully applied in similar meta-analysis studies (Jefferey et al., 2016). Equation *(2)* was used to calculate Hedge's *d* (Scheiner and Gurevitch, 2001):

$$d_{ij} = \frac{X_{ij}^{E} - X_{ij}^{C}}{s_{ij}} J \tag{2}$$

where *d* is calculated for the *j*th study in the *i*th class, and $X_{ij}{}^{C}$ is the mean of the control, $X_{ij}{}^{E}$ is the mean of the biochar treatment, s_{ij} is the pooled standard deviation of the control and experimental groups, and *J* is applied to correct for bias due to small sample size, where as the sample size increase *J* approaches 1.

Thus,

$$s_{ij} = \sqrt{\frac{\left(N_{ij}^{E} - 1\right)(S_{ij}^{E})^{2} + (N_{ij}^{C} - 1)(s_{ij}^{C})^{2}}{N_{ij}^{E} + N_{ij}^{C} - 2}} \tag{3}$$

where $N_{ij}{}^{C}$ is the total number of observations in the control, $N_{ij}{}^{E}$ is the total number of observations in the biochar treatment, $s_{ij}{}^{C}$ is the standard deviation of observations in the control, and $s_{ij}{}^{E}$ is the standard deviation of observations in the biochar treatment.

We can then further define *J* as follows:

$$\mathrm{J} = 1 - \frac{3}{4\left(N_{ij}^{E} + N_{ij}^{C} - 2\right) - 1} \tag{4}$$

A negative *d* indicates an increase in CH_4 uptake (or decrease in release) due to biochar addition and a positive *d* indicates a decrease in uptake (or increase in release). If *d* has a zero value, then there is no difference in CH_4 flux between the control and biochar amended treatments. It should be noted that *d* can only be used to compare the effect of biochar on methane flux within a category and cannot be used to compare across different variables.

2.4 Statistical analysis

Because the data gathered for analyses in this study were highly skewed, non-parametric tests were used to determine differences in Hedge's *d* among each group within each category (Aronson and Helliker, 2010). Groups with less than two treatments were excluded from the analysis. Median Hedge's *d* of each category and 95% confidence intervals were generated using Wilcoxon test. Due to large differences of CH_4 uptake/emission between upland and paddy soils, the median Hedge's *d* was also calculated in upland and paddy soils, separately. Kernel density of *d* was plotted to illustrate the variability of biochar amendment effects among studies. Locally weighted linear regression was then used to examine trends in Hedge's *d* as a function of each continuous variable (Aronson and Helliker, 2010).

Meta-analysis results including Hedge's *d* were calculated using *stats* package in R. Kernel density and locally weighted linear regression were conducted using the *ggplot2* package in R.

3 Results

3.1 Response of CH_4 uptake and emission to biochar amendment

To determine whether addition of biochar to paddy and upland agricultural soils significantly alters CH_4 emissions, we utilized results from 268 observations to compared CH_4 flux from control soils (i.e., no biochar addition) and biochar treated soils. Overall, CH_4 flux from untreated control soils was not significantly different than biochar treated soils in general (Fig. 1a). Out of 268 total comparisons, 151 showed an increase in CH_4 emission/decrease in CH_4 uptake, 111 showed a decrease in CH_4 emission/increase in CH_4 uptake, and 6 showed no change with biochar amendment. The average Hedge's *d* was -0.84 and a median of 0.124. Variability in response to biochar increased with increasing flux (Fig. 1a). The effect of biochar addition on methane flux, as quantified by Hedge's *d*, exhibited a great variability among different studies ranging from -39.33 to 17.07 (Fig. S1-S9), where negative and positive values are indicative of decrease or increase in CH_4 flux strength upon addition of biochar to control soils, respectively. Negative values may also represent an increase in CH_4 sink strength and vice versa for positive Hedge's *d* value (Jeffery et al., 2016). Non-parametric Wilcoxon rank-sum test results indicate that biochar addition leads to a significantly lower in CH_4 flux/higher CH_4 sink strength from upland soils than from paddy soils when biochar was added ($p<0.05$) (Fig. 2a), demonstrating a difference in response of the two land uses to biochar addition. Given this initial result, we further analyzed the possibly factors contributing to the difference in CH_4 flux from biochar amended paddy soil and upland soils separately.

Out of 76 observations examining biochar addition to paddy soils, 35 showed an increase in CH_4 flux strength/decrease in CH_4 sink and 41 showed a decrease in CH_4 flux strength/increase in CH_4 uptake strength. The average Hedge's *d* for biochar addition to paddy soils is -2.24 and the median is -0.86 (Fig. 2a). Similarly, addition of biochar to upland soils did not significantly change methane flux as compared to controls ($p>0.05$) (Fig. 1c). Our dataset in corroboration with past studies shows that a much larger number of studies have been performed on upland soils than paddy soils (52% more observations). Out of 192 comparisons of upland soils, 116 show an increase in CH_4 flux strength/decrease in CH_4 sink strength, 70 show a decrease in CH_4 flux strength/increase in CH_4 sink strength, and 6 show no change with biochar amendment (Fig. 2); the average of Hedge's *d* was -0.28 and the median of Hedge's d was 0.32. Separating studies by land use type also shows that biochar addition to paddy and upland soils does not result in significantly different CH_4 emission rates in general ($p>0.05$) (Fig. 1b and 1c, respectively); these results do not indicate that biochar addition has no effect on methane flux in these soils, but rather studies with increased methane flux after biochar addition cancel out the effect of the decreased flux studies in the global correlation analysis. These results also demonstrate that comparison of observations by land use only is insufficient and a more detailed examination of individual soil, biochar, and management factors is necessary to identify the specific conditions that lead to significant changes in soil CH_4 flux upon biochar addition within each setting.

3.2 Factors affecting the response of soil CH_4 uptake/emission to biochar amendment

We hypothesized that a number of soil factors, biochar characteristics, and management practices have differentiating effect on methane flux from upland versus paddy soils (as indicated by a non-zero Hedge's *d*) (Fig. 2). We provide results for both land uses combined as the "global mean" and for paddy soils and upland soils, individually, to examine the effect of each factor on CH_4 flux.

The effect of soil, biochar, and management factors on CH_4 flux upon biochar addition is represented by the magnitude of Hedge's *d* deviation from 0 (Fig. 2). Overall, the greatest effect of biochar addition is seen when considering soil properties (Fig. 2a-c) and minimal effect is imparted by variations in management practice (Fig. 2g-i). Variations in biochar properties (feedstock, pH, pyrolysis temperature, and C/N) do not have a significant effect on CH_4 flux from upland soils (Fig. 2f), while some biochar properties (feedstock, pyrolysis temperature, and C/N) contribute to significant changes in CH_4 flux strength when added to paddy soils (Fig. 2e).

3.2.1 Soil properties

Our results show that differences in soil properties correlate with the greatest difference in CH_4 flux from biochar treated soils for both land use categories. The effect of each soil property on changes in methane flux from biochar treated upland soils is reflective of the change in flux from all land uses combined (Fig. 2a and 2c), while significantly different effect of soil properties on methane flux is seen in paddy soils (Fig. 2b). A major contribution to this effect is the fewer number of observations contributed by paddy soils as compared to upland soils. This highlights the need to isolate the land use variable when examining the impact of biochar addition on methane flux, otherwise the specific impact of paddy soils would be masked.

Soil pH had a significant effect on CH_4 flux from biochar treated paddy soils (Fig. 2b). Addition of biochar to circumneutral/slightly acidic pH soils (6-<7) resulted in increased CH_4 flux strength/decreased CH_4 sink strength, while addition to acidic (pH<6) and more alkaline (pH 7-8) soils decreased CH_4 flux strength/increased CH_4 sink strength. By contrast, addition of biochar to upland soils over a wide range of pH values showed no significant difference in CH_4 flux with Hedge's *d* values clustered around 0 (Fig. 2c).

Variations in soil organic carbon (SOC) resulted in significantly different CH_4 flux from both paddy and upland soils (Fig. 2b-c). Addition of biochar to paddy soils with 20 to >30 g kg^{-1} of SOC resulted in increased CH_4 flux strength/decreased CH_4 sink strength, while treatments with soils having lower SOC (0-20 g kg^{-1}) content resulted in significantly different CH_4 flux (i.e., decreased CH_4 flux/increased CH_4 sink) (Fig. 2b). It should be noted that only 4 studies were considered for addition of biochar to paddy soils containing SOC greater than 30 g kg^{-1}. In upland soils, application of biochar to soils with high SOC (>30 g kg^{-1}) lead to significantly higher CH_4 flux strength/decreased sink strength than application to soils with 0-30 g kg^{-1} SOC content, whereas biochar addition to soils with SOC of 0 to <20 g kg^{-1} resulted in average decrease in CH_4 flux/increased CH_4 sink.

Variations in soil total nitrogen (TN) did not significantly affect CH_4 flux from biochar treated paddy soils (Fig. 2b), where TN of <1.5 and 1.5 to 3 g kg^{-1} lead to decrease in CH_4 flux/increased sink strength to no change in flux, respectively. Biochar addition to upland soils did show significantly different responses in CH_4 flux as a function of soil TN, with increased flux/decreased sink strength from soils with TN of 1.5 to >3 g kg^{-1} and significantly lower CH_4 flux/increased sink strength in soils with TN <1.5. No studies were found examining biochar addition impact on paddy soils with TN > 3 g kg^{-1}.

Soil C/N ratio had variable effect on CH_4 flux in both paddy and upland soils upon biochar addition (Fig. 2b-c). Addition of biochar to paddy soils with C/N of 15-20 had significantly higher CH_4 flux strength/lower sink strength than paddy soils with low C/N (<10). Response of upland soils to biochar additions varied widely among the 12 studies considered, where soils with high C/N (>12) ranged from showing decrease in CH_4 flux/increased sink to increased CH_4 flux/decreased sink (maximum Hedge's *d* >5) and an average Hedge's d near 0 (no effect) (Fig. 2c). A majority of upland soil studies were performed on soils with C/N of 10-15 (122 studies) where addition of biochar led to no significant change in CH_4 flux (with a narrow range of Hedge's *d* values). Biochar addition to upland soils with C/N of 15-20 resulted in increased CH_4 flux/decreased CH_4 sink, similar to response in paddy soils with the same C/N values. Our study found that soil texture has no significant effect on methane flux when biochar is applied to either paddy or upland soils (Fig. 2b-c).

3.2.2 Biochar properties

The impact of individual biochar properties including pyrolysis temperature, C/N of the biochar, feedstock, and pH of the biochar added were also examined. In general, variations in biochar properties did not result in significantly different Hedge's d values in upland soils (Fig. 2f) with ranges of Hedge's *d* values being narrow and averaging close to 0 (no change in flux) for most biochar properties. In contrast, addition of biochar to paddy soils resulted in significantly different CH_4 flux as a function of feedstock, pyrolysis temperature, and C/N of the biochar (Fig. 2e). Using biochar produced from wood and biosolids lead to decrease in CH_4 flux/increased CH_4 sink strength, however, only 7 and 4 studies were included in each analysis respectively. It should be noted that the effect of biosolids biochar has not been test in upland soils. Addition of biochar produced under high pyrolysis temperatures (501-600°C) to paddy soils lead to a decrease in CH_4 flux/increase sink strength. Low biochar C/N (<50) resulted in decreased CH_4 flux/increased sink strength, and was not significantly different than addition of high C/N (>100) biochar. Biochar pH did not have any significant effect on CH_4 flux in paddy soils.

3.2.3 Management practice

The impact of management practice on CH_4 flux from biochar treated soils was examined by separating studies into categories including experimental time (number of days of the experiment), experimental method (field-based, pot experiments, and incubation experiments), and fertilizer application rate (nitrogen, phosphate, and potassium application). Overall, variations in management practices generally did not have a significant impact on CH_4 flux from biochar treated soils (Fig. 2g-i). For paddy soils, fertilizer application rate of 5-10% (only two studies found), shorter experimental time (<60 days), and no fertilizer application (phosphate and potassium application) were associated with significantly lower CH_4 flux. Other management practices were not associated with significant change in CH_4 flux with biochar treatment. Methane flux from upland soils were essentially unchanged as a function of various management practices, aside from a slight increase in CH_4 flux in treatments with low potassium application rate (<150 kg ha^{-1}).

3.3 Locally weighted linear regression and linear additive model

The effect of a number of continuous variables on methane dynamics was visualized using locally weighted linear regression (*lowess* function in R) (Fig. 3 and Fig. S10, S11). Through this visualization, we are able to have a more thorough and accurate view of the quantitative relationship between change in methane flux (Hedge's *d* values) as function of each soil, biochar, and management variable continuously, rather than just as an average Hedge's *d* bracketed by a range as presented in Fig. 2. Using these local regression functions, we can approximate the relationship between methane flux/sink and a given variable's range of values; subsequently, a weight value is determined from quantitative relationship within each of these regions, which can then be applied to the variable during model parameterization. Our visualization shows that for many soil properties, there are ranges of soil property variable values that correlate with Hedge's *d* (Fig. 3). For model parameterization, we are most concerned with variable regions that lead to Hedge's *d* values that are significantly different than 0, which are identified as where the red local regression function lies outside of the confidence interval delineated by the gray zone. We provide here a summary of the notable variable ranges for which biochar addition significantly changed CH_4 flux. For biochar addition to all land uses, soil organic carbon (<20 g kg^{-1}) and total nitrogen (<2.0 g kg^{-1}) are associated with negative Hedge's *d* (Fig. 3a and 3b). Soil clay (<10%) is associated with negative Hedge's *d* (Fig. 3e) and clay (10-20%) is associated with positive Hedge's *d* in biochar treatment upland soil (Fig. 3o). Biochar treatment of upland soils with C:N of 8 to 12 is associated with a negative Hedge's *d* (Fig. 3m). Biochar pyrolysis temperature (>500°C) and C/N (<70) in biochar treatment paddy soil is correlated with negative Hedge's *d* (Fig. S10). Management practices were not significantly associated with non-zero Hedge's *d* values upon biochar treatment of either land use type (Fig. S11).

4 Discussion

Past studies have reported contrasting effects of biochar addition on CH_4 flux from soils, where addition of biochar have lead to significant increase, decrease, or no change in CH_4 flux from various agricultural soils (Jeffery et al., 2016; Song et al., 2016). We hypothesized that land use (paddy versus upland) in combination with specific soil properties determined whether increased CH_4 production or increased CH_4 sink strength would result upon biochar addition. We first examined the contribution of land use by comparing results from biochar addition to paddy versus upland soils and found that such a global analysis results in what appears to be unchanged CH_4 flux as compared to untreated soils (Fig. 1). This lead to a more detailed examination of the specific soil, biochar, and management factors that have been shown to contribute to changes in CH_4 flux (Lehmann et al., 2011). Our meta-analysis reveals that many factors can affect CH_4 flux response significantly with biochar input, in particular, soil properties such as SOC, TN, C/N, pH, biochar properties including pyrolysis

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temperature and C/N, application rate, and experimental design and management practices including length of experiment, experimental methods, and P and K fertilizer application rate (Fig. 2).

Net soil CH_4 emission is determined by a complex set of biogeochemical processes occurring simultaneously including the competition between methanogenic and methanotrophic process (Schink, 1997). Methanogenesis can be stimulated or inhibited by changes in soil moisture, soil redox state, soil pH or the differences in adsorbed organic compounds and inorganic constituents (Wang et al., 1993; Yang and Chang, 1998). Studies have shown that microbial substrate availability strongly affects CH_4 production (Christensen et al., 2003) where CH_4 production can be inhibited by low soil organic matter content (Xie et al., 2013). Biochar addition to paddy and upland soils with low SOC (0-15 g kg^{-1}) lead to decreased CH_4 flux/increased sink strength indicating that methanotrophs likely out compete methanogens under limited SOC conditions, regardless of redox conditions. It should be noted that a negative Hedge's *d* value doesn't not necessarily indicate a change in flux direction, therefore, incorporation of biochar could enhance aerobic conditions that lead to an increase CH_4 sink. In contrast, high SOC (>20 g kg^{-1}) and soil C/N (15-20) are associated with significant increase in CH_4 flux (positive Hedge's *d* values) (Fig. 2b-c). This suggests that methanogenesis is enhanced in both paddy and upland soils upon biochar addition above a SOC concentration threshold. Biochar amendment to soils has been shown to provide additional habitats for microbes (Mukherjee and Lal, 2013) and can adsorb and retain organic substrates from the soil matrix to fuel microbial processes (Wardle et al., 2008); however, it is unlikely that biochar C is directly respired due to its recalcitrance (Knoblauch et al., 2011; Xie et al., 2013). Biochar addition can also increase soil moisture (Wang et al., 1993; Yang and Chang, 1998), which may promote the growth of methanogens particularly in combination with abundantly available SOC, while potentially lead that methanogens out compete methanotrophs within upland soils.

Variations in soil pH was also significantly affected how paddy soils responded to biochar additions, but no significant change in flux was seen for a range of upland soil pH (Fig. 2a-c). Optimal growth of methanogens is usually achieved under near neutral or slightly alkaline pH conditions (Garcia et al., 2000), while optimal range for methanotrophy includes slightly acidic conditions (Le Mer and Roger, 2001). Biochar input has been shown to increase soil pH owing to its alkaline characteristic (Chan and Xu, 2009; Liu et al., 2011; Van Zwieten et al., 2015). Inundation of paddy soils leads to accelerated weathering of soil minerals toward poorly crystalline oxides including aluminum oxides. Under acidic soil conditions, aluminum can be ready available in soluble form as Al^{3+} which can have toxic effects on both plants and microbes, with methanotrophs being more sensitive to high Al^{3+} concentrations (Tamai et al, 2007). Therefore, increased pH due to biochar addition removes Al^{3+} through precipitation of Al hydroxides. However, this process would be expected to have equal effect in upland soils. A distinction between the two land uses then is the action of biochar integration into flooded soils can help aerate the soil temporarily and can also impart aerobic microsites where methanotrophy is promoted. This effect would be emphasized for acidic soils because of the compounding effect of Al^{3+} removal and partial aeration of soil microsites.

In contrast to acidic soils, application of biochar to near neutral soils (pH of 6-7) is associated with increased CH_4 flux in paddy and upland soils (Fig. 2b-c), possibly due to lower tolerance of methanotrophs to pH shifts from neutral conditions (Jeffery et al., 2016). The kernel density estimation of biochar pH from all studies considered shows that most biochar applied to soils were between pH of 8-10 (Fig. S4-6). Unfortunately, there is insufficient data within our study to determine whether a relationship between soil pH change and biochar amendment exists, making it difficult to determine whether the positive relationship between biochar amendment and CH_4 flux is due to soil pH change. However, our results demonstrate that biochar amendment to circumneutral pH soils likely will increase CH_4 flux.

Many studies have reported that the total porosity and aeration of soils are increased with biochar input, leading to change in oxygen and water status (Lehmann et al., 2011; Mukherjee and Lal, 2013). However, our meta-analysis shows that variations in soil texture had a non-significant effect on CH_4 release/uptake with biochar input in either paddy or upland soils (Fig. 2a-c). Addition of biochar to Higher clay content in both upland and paddy soils appear to be associated with slightly higher CH_4 flux/decreased sink relative to other soil textures, where addition of biochar course texture is associated with decreased flux/increased sink strength for both land uses. This result may indicate that addition of biochar to coarser soil textures, which are already expected to favor methanotrophy particularly in upland soils, further enhances methanotrophs or inhibits methanogens by providing even greater aeration and microsites. Again, the lower number of studies on paddy soils also restricts our confidence in defining possible driving mechanisms. Further investigation into this counterintuitive result is needed to clarify the role of soil texture in influencing methane flux from biochar amended paddy and upland soils.

Though soil properties appear to have the most influence on change in CH_4 flux with biochar amendment, biochar feedstock, pyrolysis temperature, and biochar C/N were also associated with significant changes in CH_4 flux change (Fig. 2d-f). However, due to the low number of studies using biosolids (n = 4) and wood (n = 9), the results may not be conclusive. Biochar heat treatment temperature is a major factor influences pyrolytic production and functional group composition of the resulting biochar (Keiluweit et al., 2010). Biochar produced at 400-700°C have a large amount of quinone and hydroquinone moieties, which can act as sorbents of electron donors and acceptors (Klüpfel et al., 2014; Zhou et al., 2014) that are metabolized during methanogenesis (Saquing et al., 2016). Application of biochar produced at 500-600°C significantly decreased CH_4 flux from paddy soils, but not upland soils, possibly due to an inhibitory effect of certain functional groups on methanogenesis under anaerobic conditions. Further research is needed to better understand how chemical differences in biochar surface due to pyrolysis temperature affects metabolic processes of methanogens and methanotrophs in upland and paddy soils.

The correlation of low biochar C/N (<50) with decreased CH_4 flux/increased CH_4 sink may be due to inhibition of methanogenesis by increase in soil nitrate upon biochar addition. Nitrate has been shown to inhibit methane production due to greater energetic yield provided by denitrification process, allowing nitrate reducing bacteria to outcompete methanogens for electron donors such as acetate (Chidthaisong and Conrad, 2000). Additionally, denitrification produces toxic intermediates (i.e., NO_2^-, NO, N_2O), the presence of which have been show to inhibit the growth of acetoclastic methanogens (Scheid et al., 2003; Klüber and Conrad, 1998) .

Aside from inhibition of methanogenesis by the presence of metabolic intermediates, decreased methane flux upon biochar addition to low C/N soils may also be caused by high ammonium and nitrate concentrations from N fertilizer additions in those studies. This would lead to decreased organic matter decomposition and decrease in substrate access by methanogens (Lucas and Casper, 2008). However, our results showed N fertilizer application rate have no significant influence on methane flux upon biochar input (Fig. 2g-2i), which has also been observed in other studies (Neff et al., 1994).

Variations in most management practices do not result in a significant change in methane flux upon biochar addition to upland soils, in contrast to paddy soils where management practice appears to alter flux significantly in multiple categories. Biochar amendment to soils without P and K fertilizer application appears to inhibit CH_4 production/increase sink strength in paddy soil (Fig. 2h). Phosphate availability can be enhanced with alkaline biochar addition in acidic soils by increasing soil pH (Nelson et al., 2011; Novak et al., 2009). Potassium availability generally also increases as soil pH increases and becomes optimally available at pH above 6.5 (Subedi et al., 2016). Biochar amendment to soils may have increased the P and K availability, allowing methanotrophs to outcompete methanogens the growth of which have been shown to be inhibited in the presence of phosphate (Paulo et al., 2005). A notable finding from our meta-analysis is that experimental time and method (field, pot, or incubation) can also significantly influence methane flux from paddy soils. This demonstrates that experimental design must be carefully considered when interpreting the results from biochar amendment studies and that when possible, various scales of experiment methods (e.g., field versus pot) should be included in a single study for cross-study comparisons.

In order to build linear additive models that can be used to predict soil methane flux in response to biochar additions, variables to be used in model matrix need to be evaluated based on the local linear regression results. By individually evaluating each variable using a continuous regression function, we are able to identify the specific range of parameter values that result in increase versus decrease in methane flux/change in sink strength (Fig. 3). Our meta-analysis results emphasize the need to integrate land use type, soil properties, and a select number of biochar properties, with weaker emphasis on management practices variables when building additive models. For example, by excluding management practice parameters, the model goodness-of-fit will likely increase while also decreasing computational time (Wood, 2006).

5 Conclusions

Biochar addition to agricultural soils have been shown to result in highly variable change in methane flux, from increased methane emissions/decreasing sink strength of the soil, decreased emissions/increase in sink strength, to having no effect. Here, we demonstrate that more detailed examination of land use, soil, and biochar characteristics an help define parameters that significantly affect soil response to biochar amendment. Although biochar amendment does not appear to influence the CH_4 fluxes in soil ecosystems when a global mean is calculated, we demonstrate that paddy and upland soils have significantly different responses to biochar addition particularly when soil parameters are taken into consideration individually. We demonstrate the use of local regression to generate a continuous function that provides more detailed information into the quantitative ranges of conditions that lead to increased/decreased methane flux or increased/decreased methane sink strength. We also show that variation in management practice have no significant effect and therefore should be removed when building additive predictive models to increase model goodness-of-fit. The incorporation of these findings into global climate models may yield a more accurate representation of the interactions between biochar input and carbon cycles, and better predictions of future climatic feedback trends.

Author contributions. WC performed data collection and data anlaysis, SCY performed data interpretation. The manuscript was written by WC and SCY with comments from JM.

▪ *Competing interests. The authors declare that they have no conflit of interest.*

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Figure captions

Fig. 1. The relationship between methane flux in control (no biochar addition) and treatment (biochar added) plots. Negative flux values indicate atmospheric methane uptake by soils. The red line is a local regression with 95% confidence interval shown in gray. The black line is the 1:1 line for comparison.

Fig. 2. The effect of soil properties, biochar properties, and management factors on methane flux upon biochar amendment as indicated by Hedge's *d.* Points indicate median values bounded by 95 % confidence interval. Negative *d* values indicate decreased methane emission/increased methane sink by soils with biochar addition. Sample size is indicated in parentheses. Different letters indicate statistically significant differences between median (Wilcoxon rank-sum test, p<0.05). The dotted vertical line indicates Hedge's *d* of 0 or no change in methane flux upon biochar addition.

Fig. 3. The relationship between the soil properties and Hedge's *d* for all soils (a-e), paddy (f-j) and upland (k-o) soils. Shaded bands indicate 95% confidence intervals for the mean of each treatment. Negative *d* values indicate decreased methane emission/increased methane sink by soils with biochar addition. The dotted vertical line indicates Hedge's *d* of 0 or no change in methane flux upon biochar addition.

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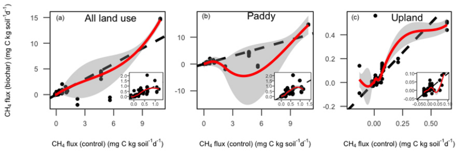

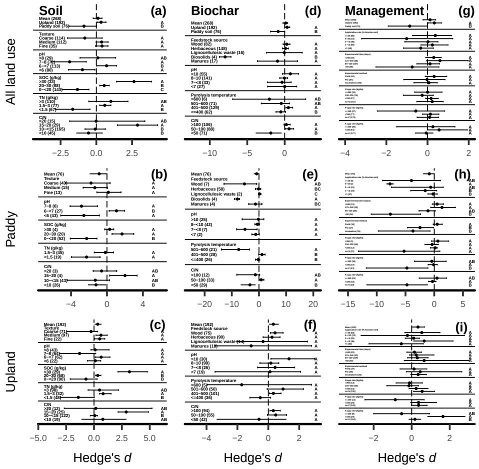
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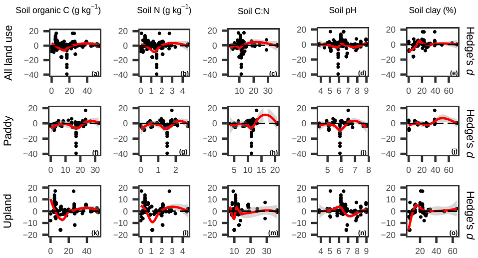
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