Ecosystems (2018) 21: 1692–1705
https://doi.org/10.1007/s10021-018-0248-y

ECOSYSTEMS

Variation in Soil Methane Release or Uptake Responses to Biochar Amendment: A Separate Meta-analysis

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Abstract

Agricultural soils play an important role in the atmospheric methane (CH_4) budget, where paddy soils can contribute significant CH_4 to atmosphere whereas upland soils may act as a source or sink of atmospheric CH_4, dependent on soil water conditions. Biochar amendments have effects on soil CH_4 production or oxidation processes in individual experiments, but the causative mechanisms are yet to be fully elucidated. To synthesize the response of soil CH_4 release or uptake to biochar amendment, we performed a meta-analysis using data from 61 peer-reviewed papers with 222 updated paired measurements. When averaged across all studies, biochar amendment significantly decreased CH_4 release rates by 12% for paddy soils and 72% for upland soils, and CH_4 uptake rates by 84% for upland soils. Neither soil CH_4 release nor uptake responses to biochar amendment were significant in field soils. Nitrogen (N) fertilizer application would weaken the response of soil CH_4 release or uptake to biochar amendment. Biochar-incurred decreases in soil CH_4 release and uptake rates were the largest in medium-textured soils or neutral-pH soils. Soil CH_4 release or uptake responses to biochar were also significantly altered by biochar characteristics, such as feedstock source, C/N ratio, pH, and pyrolysis temperature. The results of this synthesis suggest that the role of biochar in soil CH_4 mitigation potential might have been exaggerated, particularly in fields when biochar is applied in combination with N fertilizer.

Key words: biochar; methane; climate change; paddy soil; upland soil; meta-analysis.

Received 19 December 2017; accepted 16 March 2018;
published online 9 April 2018

Electronic supplementary material: The online version of this article (https://doi.org/10.1007/s10021-018-0248-y) contains supplementary material, which is available to authorized users.

Cheng Ji, Yaguo Jin have contributed equally to this work.
Author Contribution: JWZ and SWL conceived the investigation. SWL, JC, KY, CL and CJ extracted the data from literature and constructed the database. SWL, DLK and YGJ performed the statistical analyses. JWZ and SWL wrote the manuscript.

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Introduction

Methane (CH_4) is one of the most potent long-lived atmospheric greenhouse gases (GHGs) that threatens earth's climate system by exerting a relative sustained flux global warming potential (SGWP) 45 times greater than that for carbon dioxide (CO_2) on a mass basis over the 100-year time horizon (IPCC 2013). Since the pre-industrial era, the atmospheric

CH_4 concentration has increased from 700 ppbv to almost 1800 ppbv (Dlugokencky and others 2009). Atmospheric CH_4 is primarily derived from biological sources mediated by methanogenesis under anaerobic conditions, which accounts for more than 70% of the global total (Denman and others 2007). Soils play an important role in atmospheric CH_4 budget because paddy soils can release significant CH_4 to the atmosphere, whereas upland soils can uptake atmospheric CH_4, acting as an important source or sink of atmospheric CH_4, respectively (IPCC 2013).

Biochar mainly derived from pyrolyzing biomass has been increasingly proposed as a potential management strategy to improve crop productivity and soil quality (Glaser and others 2002; Chan and others 2007; Laird 2008; Woolf and others 2010; Lehmann and others 2011; Case and others 2014). It has also been proposed to be a potential alternative for enhancing soil C sequestration and/or mitigating soil GHGs emission (Lehmann 2007; Stewart and others 2012; Zhang and others 2013; Li and others 2014). By summarizing available data, prior meta-analysis studies showed that biochar amendment can significantly enhance soil C sequestration and reduce nitrous oxide (N_2O) emissions from soils (Cayuela and others 2013; Sagrilo and others 2015; Liu and others 2016). Recently, several studies have addressed the response of soil CH_4 fluxes to biochar amendment across soils using meta-analysis procedures (Jeffery and others 2016; Song and others 2016; He and others 2017). Unfortunately, these synthesis studies failed to partition biochar effects on CH_4 release from paddy soils and CH_4 uptake of upland soils, although soil CH_4 production and oxidation are two quite different processes ultimately determining the net soil-atmosphere CH_4 balance.

Increasing evidence on CH_4 fluxes from biochar-amended agricultural soils has generated inconsistent results (Spokas and Reicosky 2009; Rogovska and others 2011; Zhang and others 2010, 2012a, b, 2013). For instance, soil CH_4 release rates were almost completely suppressed in biochar-amended acidic soils in the Eastern Colombian Plains (Rondon and others 2005), in contrast to no significant effects of biochar in a calcaric fluvisol (Knoblauch and others 2008), or even an increase in soil CH_4 release rates due to biochar amendment in a paddy soil (Zhang and others 2010). Similarly, soil CH_4 uptake rates were found to increase (Karhu and others 2011; Scheer and others 2011; Zhang and others 2012a) or decrease (Borchard and others 2014; Hawthorne and others 2017) following biochar amendment into upland cropping soils. Nevertheless, these increasing individual experimental studies allow us to use meta-analysis to reexamine these ongoing inconsistencies and elucidate the causative mechanisms.

A number of hypotheses have been proposed to explain the effects of biochar on soil CH_4 fluxes. First, biochar amendment can improve soil aeration, leading to a decrease in soil CH_4 release or an increase in CH_4 oxidation, especially in soils without waterlogging (Zhang and others 2016). Second, the response of soil CH_4 release or uptake to biochar amendment may be subjected to the amount and form of soil organic matter, soil N availability and even the interaction between them (Karhu and others 2011). Third, the structural and chemical properties (for example, feedstock, pH, C/N ratio, and pyrolysis temperature) of biochar itself can also constitute the potential driving factors affecting methanogenic or CH_4 oxidation activities in soils (Spokas and Reicosky 2009). Therefore, soil CH_4 release or uptake responses to biochar amendment need to be separately analyzed using meta-analysis, which would help to gain an insight into the effect of biochar on soil CH_4 fluxes, in terms of its source or sink potential of atmospheric CH_4.

Although numerous individual experiments have been conducted to test the effectiveness of biochar on mitigating soil CH_4 emissions in agricultural soils, the full range of mechanisms and consequences behind these effects remain poorly elucidated. In particular, there is currently a lack of systematic synthesis because biochar performance in soils on CH_4 varied across sites due to environmental and management factors (for example, soil properties, biochar sources, climates or management practices). Besides, the response of soil CH_4 release or uptake to biochar amendment may substantially differ across soils based on the two intrinsic factors conflicting soil CH_4 production and oxidization processes. Meta-analysis has been developed for quantitative integration of results from individual studies, which is increasingly used in studies with respect to ecological issues and issues on greenhouse gases mitigation (Knorr and others 2005; Akiyama and others 2010; Van Groenigen and others 2011; Shan and Yan 2013).

Here, we compiled 222 updated paired measurements derived from 61 peer-reviewed papers to synthesize the response of soil CH_4 release or uptake to biochar amendment using meta-analysis procedures. The main objective of this study was to quantitatively and separately examine the response of soil CH_4 release or uptake to biochar amendment, and simultaneously to address earlier review limitations. We also attempted to identify the key factors driving the response of soil CH_4 fluxes to biochar amendment. This meta-analysis study would help to achieve a scientific assessment of the practical role of biochar in regulating soil-atmosphere CH_4 exchange balance.

Methods

Data Extraction and Compilation

We conducted a detailed review of the literature reporting the effects of biochar amendment on soil CH_4 fluxes prior to October 2017. This included a keyword search in Web of Science (ISI) and Google Scholar (Google Inc.), and papers published in the China Knowledge Resource Integrated Database (CNKI) with English abstracts. Different combinations of search keywords (''biochar'' OR ''charcoal'' OR ''black carbon'' AND ''methane'' OR ''CH_4'' AND ''soil'') were used for data extraction. Eventually, data were collected from 61 published research studies with 222 individual paired measurements by integrating both the control and biochar-amended treatments (Supporting Information, Appendix S1 and S4).

For each paired measurement, we gathered a range of original documented information by integrating mean and/or cumulative CH_4 fluxes, standard deviation (SD), and number of replicates from both biochar amendment and control treatments. Meanwhile, actual or original land-use type, soil properties (for example, texture and pH), biochar source and characteristics (feedstock, pyrolysis temperature, pH, C/N ratio), biochar application rate and experimental condition (field/pot/incubation study, rate and source of fertilizer, and duration) were also included when available. Throughout the text, the term of positive and negative metrics represents biochar-induced increase and decrease in soil CH_4 release or uptake rates, respectively.

In further data compiling prior to meta-analysis, we categorized the soils into upland and paddy soils based on the actual or original land-use type and the database established, where the datasets from grassland and forest were integrated into the upland grouping category to calculate the effect size in this analysis. Soil texture was grouped into three basic classes (that is, fine, medium and coarse) due to the inconsistent reports of soil texture in the literature (for example, general qualitative description, particle size distribution, soil taxonomical unit). Based on the information on soil layer and bulk density (BD) of soils presented in the literature, we uniformly transformed the biochar rates into area-based amounts (expressed as t ha^{-1}). When cumulative CH_4 fluxes failed to be directly obtained, we estimated the value by multiplying the average daily fluxes with the experimental days. When data were presented in only graphical form, they were digitized using the software plot digitizer version 2.6.2 to extract the data points (Cayuela and others 2013). All data were subjected to a standardization process to allow for comparisons.

Besides field studies, the studies under controlled experimental contexts (laboratory incubation or pot studies) were also introduced to fully evaluate the integrative effect of biochar on CH_4 fluxes across soils (Figure 1). For incubation or pot studies, however, only those highly simulating the field water capacity (30–85% WFPS) were included in this analysis to guarantee the ability to group the available soils into different original land-use types. Only studies with at least three replicates were included for this analysis. However, about 30% (56 of 222) of the measurements failed to report any information on variance (SD, standard error, or variance). In these cases, efforts were made to obtain these from the corresponding authors. Otherwise, we assigned the averaged SD for missing values within the given sub-grouping category to include as many studies as possible in this study.

Meta-analysis

In the database, all paddy soils and 83 paired measurements of upland soils exhibited positive CH_4 fluxes (referring to soil CH_4 release) and biochar amendment did not change the source role of atmospheric CH_4. About 62 paired measurements of upland soils consistently showed negative CH_4 fluxes (referring to soil uptake of CH_4) for both biochar treatments and controls, acting as the sink role of atmospheric CH_4, and thereby the absolute values of negative CH_4 fluxes were adopted for effect size calculation to avoid making $\ln R$ problematic during meta-analysis. Seven paired measurements showed a shift from source to sink of atmospheric CH_4 following biochar amendment, and five paired measurements showed a shift from sink to source of CH_4 in grassland and forest soils, which finally led to the exclusion of them from this analysis to allow for solid performance of meta-analysis procedures.

We calculated effect size based on the natural log-transformed response ratio ($\ln R$) using the means of cumulative CH_4 fluxes from biochar amendment (X_t) and control (X_c) groups across

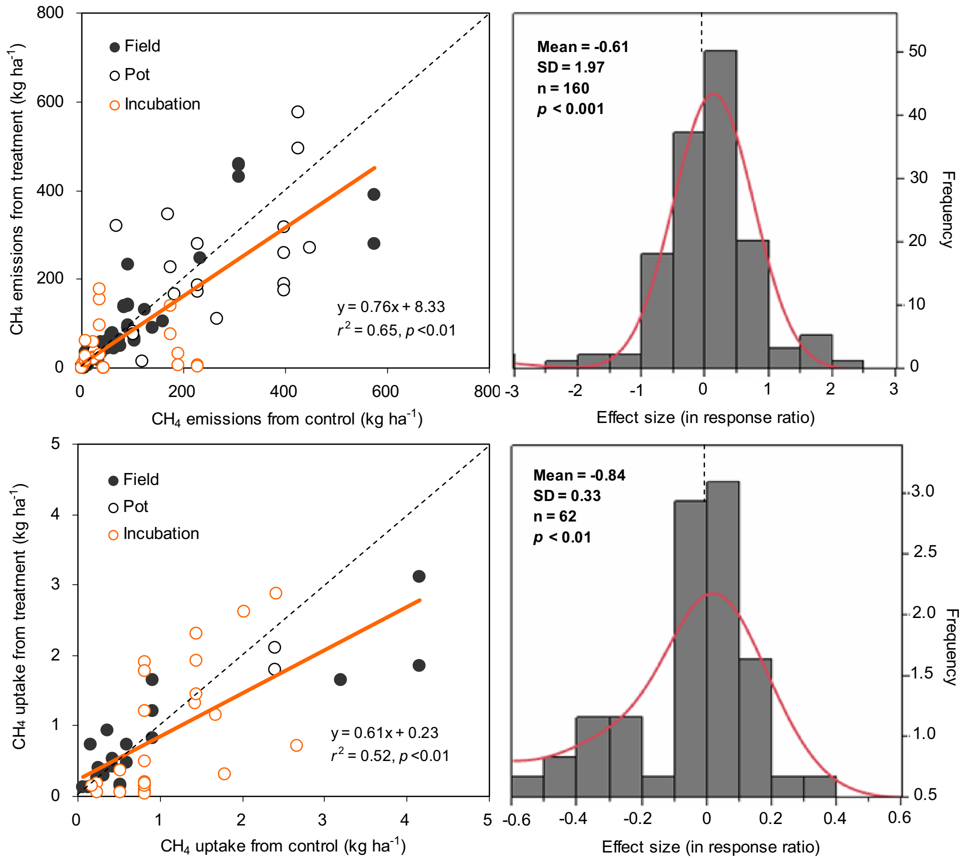

Figure 1. Frequency distribution of effect sizes among all studies and correlation of soil CH_4 release or uptake in biochar-amended treatments against those of controls (with a mean and SD of − 0.61 and 1.97 for CH_4 release data and − 0.84 and 0.33 for CH_4 uptake data, respectively). The dotted line represents the theoretical 1:1 line where the release or uptake rate of CH_4 from amended and control soils is equal, whereas the solid line represents the linear regression for all individual observations. The regressions for CH_4 release and uptake have a strength of R^2 = 0.65 and 0.52, respectively.

soils. The standard deviations of both biochar treatment and control were included as a measure of variance:

$$\ln R = \ln(X_t/X_c) = \ln(X_t) - \ln(X_c) \qquad (1)$$

where X_t and X_c are means in the treatment and control groups exposed to biochar present and absent, respectively. Its variance (v) is estimated as:

$$v = \frac{s_t^2}{n_t x_t^2} + \frac{s_c^2}{n_c x_c^2} \qquad (2)$$

where n_t and n_c are the sample sizes for the treatment and control groups, respectively; s_t and s_c are the standard deviations for the treatment and control groups, respectively.

The categorical random effects model was used to calculate mean effect size for each grouping category with a weighted meta-analysis approach. Groups with less than two treatments were excluded from the analysis. Mean effect sizes of each category and the 95% confidence intervals (CIs) generated by bootstrapping (9999 iterations) were calculated with the mixed-effect model by R (version 3.2.0, R Development Core Team 2016). Actually, all the original mean effect sizes (ln$R$) were further normalized by adding one to make the line one in the Figures as reference where biochar has no effect on CH_4 fluxes relative to controls being equivalent to 1, which were considered to be significantly different from those of X_c if the 95% CIs did not overlap with line one (ln$R$), and significantly different from one another if their 95% CIs did not overlap (Figures 2, 3, 4, 5, 6). To further address the differences among sub-grouping categories, between-group heterogeneity (Q_b) was examined across all data for a given response variable.

We examined publication bias using the funnel plot and Egger regression (Jennions and others

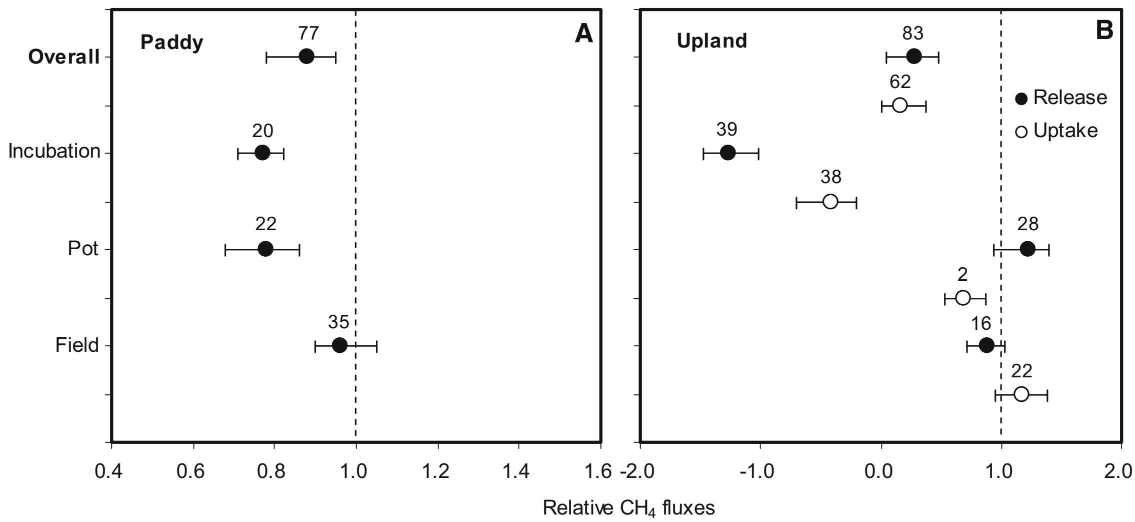

Figure 2. Response of soil CH_4 fluxes to biochar amendment differed with land-use type and experimental method, shown as relative fluxes (CH_4 release or uptake rate from controls are equal to one). Mean effect and 95% CIs are shown. Numerals indicate number of observations. 'Overall' indicates the integrated effect across experimental methods. The datasets from grassland and forest were integrated into the upland grouping category. The closed and open symbols represent original control soils acting as source or sink of CH_4, respectively. For upland pot studies, no available soils acted as sinks of CH_4.

2013). The funnel plot is a scatterplot of the effect sizes against their standard errors and the absence of publication bias can be achieved providing that the datapoints distribute symmetrically in a ''funnel'' shape around the mean effect size (Appendix S2). The potential symmetry of the funnel plot was further assessed using the Egger's regression model (Appendix S3). The frequency distribution of ln*RR*++ was plotted to reflect variability of individual measurements and test the normality of all datasets. The normality examination was carried out using JMP version 7.0 software.

Results

Soil CH_4 Fluxes Response to Biochar Amendment

By separating soil CH_4 release and uptake data, significant positive linear relationships were observed between soil CH_4 fluxes in biochar-amended treatments against those in controls, with a strength of $R^2 = 0.65$ and 0.52, respectively (Figure 1). Frequency distribution of overall effect sizes across all studies tended to have a normal distribution with a mean and standard deviation of − 0.61 and 1.97 for CH_4 release data and − 0.84 and 0.33 for CH_4 uptake data, respectively, suggesting that the datasets were relatively homogenous (Figure 1). When averaged across all studies, biochar amendment significantly decreased soil CH_4 release and uptake rates by 61% (confidence interval, CI − 81 to − 47%) and 84% (CI − 102 to − 69%), respectively. In addition, the existence of publication bias was not suggested by the funnel plot and Egger regression for the both CH_4 release and uptake data across all experimental methods and land-use types (Appendix S2 and S3).

Land-Use Type and Experimental Method

Soil CH_4 fluxes response to biochar amendment depended on land-use type and experimental method (Table 1 and Figure 2). When averaged across all studies in paddy soils, biochar amendment significantly decreased CH_4 release rates by 12% (CI − 26 to − 3%), largely attributed to incubation and pot studies (Table 1 and Figure 2). Relatively, paddy soils showed the weakest negative response (− 4%, CI − 6 to 11%) in field studies. When pooling data on upland soils acting as a source of atmospheric CH_4, biochar amendment significantly decreased CH_4 releases from upland soils by 72% (CI − 97 to − 44%), which was overwhelmingly contributed by incubation studies (− 226%, CI − 243 to − 191%). When averaged all studies on upland soils acting as a sink of CH_4, biochar amendment consistently decreased soil CH_4 uptake in both incubation and pot studies,

Table 1. Between-group Variability (Q_b) Among Observations (n) Suggesting their Potential as Predictor Variables Influencing the Responses of Soil CH_4 Release or Uptake to Biochar Amendment

Categorical variables	Soil CH_4 release				Soil CH_4 uptake			
	n	Q_b	P	df	n	Q_b	P	df
All studies	160	–	–	–	62	–		–
Land-use type	160	0.32	0.02	5	–	–	–	–
Experimental method	160	4.37	ns	5	62	11.42	0.01	3
Soil texture	158	30.45	< 0.001	5	62	12.06	0.01	5
Soil pH	158	28.76	< 0.001	5	62	32.51	< 0.001	3
Fertilizer type (kg N ha^{-1})	84	12.10	< 0.001	4	22	13.24	< 0.001	3
Biochar applied rate (t ha^{-1})	160	9.65	0.01	5	61	14.10	< 0.01	2
Feedstock source	160	21.56	< 0.01	5	61	23.11	< 0.001	4
Pyrolysis temperature (°C)	157	10.23	< 0.01	3	52	12.49	< 0.001	3
Biochar pH	143	6.12	0.02	3	42	3.48	0.04	2
Biochar C/N ratio	155	31.57	< 0.001	5	62	1.23	ns	3

A larger Q_b is a better predictor of variation than a variable with a smaller Q_b; df refers to degrees of freedom.
Statistical significance at the $P < 0.05$ possibility level; ns, not statistically significant.

but slightly increased soil CH_4 uptake in field studies, leading to an overall significant decrease by 84% (CI − 102 to − 69%, Figure 2).

Soil Texture and pH

The response of CH_4 fluxes to biochar amendment depended on soil texture (Table 1 and Figure 3). For soils acting as a source of CH_4, significant responses of soil CH_4 release to biochar were positive in soils with fine texture (+34%, CI 8–62%), in contrast to negative responses in soils with medium (− 170, CI − 214 to − 126%) or coarse texture (− 32%, CI − 78 to − 12%; Figure 3A). For upland soils as a sink of CH_4, soil CH_4 uptake rates were significantly decreased following biochar amendment over all investigated soil textures and the strongest negative response occurred in soils with medium texture (− 129, CI − 144 to − 78%).

Across all observations, experimental soils were divided into acid, neutral, and alkaline soils, corresponding to soil pH < 6.5, 6.6–7.5, and > 7.5, respectively. For soils acting as source of CH_4, biochar significantly decreased soil CH_4 release rates by 128% (CI 96–142%) in neutral soils, in contrast to a less decrease in alkaline soils or significant increase in acid soils (Table 1 and Figure 3B). For upland soils acting as sink of CH_4, biochar amendment significantly decreased CH_4 uptake rates both in acid and neutral soils. The decrease in CH_4 uptake rates was the greatest in neutral soils (− 1640, CI − 185 to − 122%), against minor positive responses in alkaline soils (+11, CI − 3 to 27%).

Combined Effect of Biochar with Fertilizer

The response of soil CH_4 fluxes to biochar amendment was altered by fertilizer under biochar combined with fertilizer application conditions (Figure 4). Biochar amendment significantly decreased soil CH_4 release rates by 115% (CI 99–131%) for unfertilized soils, as a contrast to a minor decrease in soil CH_4 release for fertilized soils (− 9%, CI − 14 to 1%). Similarly, the biochar-induced decrease in CH_4 uptake of upland soils was weakened by fertilizer application (Figure 4). The response of soil CH_4 fluxes to biochar amendment also differed with fertilizer type (Figure 4). Biochar combined with synthetic and organic N fertilizer led to a decrease by 39% (CI 26–47%) and 8% (CI 2–13%) in CH_4 release rates, respectively, whereas biochar combined with compound N fertilizer incurred a significant positive response (+28%, CI 22–39%). The datasets for upland soils acting as a sink of CH_4 were only available within the subgrouping category of synthetic and organic N fertilizer types in this analysis, showing significant negative effects on soil CH_4 uptake for synthetic N (− 25%, CI − 41 to − 17%) but positive effects for organic N fertilizer (+20%, CI 18–23%).

Biochar Application Rate

The response of soil CH_4 fluxes to biochar amendment depended on application rate of biochar (Table 1 and Figure 5). The response of soil CH_4 release to biochar amendment generally decreased with biochar rates, shifting from the highest posi-

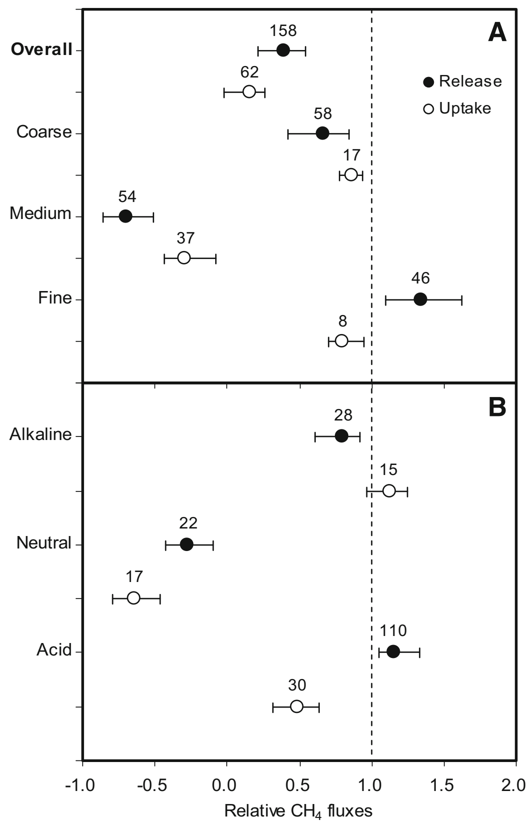

Figure 3. Response of soil CH_4 fluxes to biochar amendment dependent on soil texture (**A**) and pH (**B**), shown as relative fluxes (CH_4 release or uptake rate from controls are equal to one). Mean effect and 95% CIs are shown. Numerals indicate number of observations. 'Overall' indicates the integrated effect across different soil textures. All datasets were included in this meta-analysis category. The closed and open symbols represent original control soils acting as sources or sinks of CH_4, respectively.

tive response at rates less than 20 t ha^{-1} (+28%, CI 24–37%) to the lowest negative response at rates of over 60 t ha^{-1} (− 186%, CI − 241 to − 152%). Similarly, for upland soils acting as a sink of CH_4, the response of soil CH_4 uptake rates to biochar also presented a declined trend with biochar rates, from the insignificant increase effect (+9%, CI − 1 to 13%) at the amended rate lower than 20 t ha^{-1} to the largest decrease effect (− 171%, CI − 222 to − 143%) at an amended rate over 60 t ha^{-1} (Table 1 and Figure 5).

Biochar Leading Characteristics

The response of soil CH_4 fluxes to biochar was subjected to its leading characteristics such as feedstock source, C/N ratio, pH and pyrolysis temperature. Most studies have been undertaken using wood (64 datasets, 28% of total) and herbage (125 datasets, 56% of total) as biochar-derived materials, whereas only a small number of studies used manure (8 datasets), biowaste (7 datasets) and ligneous (17 datasets) materials as biochar feedstock sources (Figure 6A). For soils acting as a source of CH_4, the response of soil CH_4 release to biochar differed significantly among biochar material sources (Table 1 and Figure 6A). Significant negative responses were found to wood, biowaste, ligneous and herbage materials as feedstock sources, contrary to significant positive responses to manure-derived biochar (+21%, CI 19–28%). For upland soils as a sink of CH_4, soil CH_4 uptake rates were significantly decreased across wood, ligneous and herbage feedstock materials, whereas slightly increased using manure as feedstock sources (+8%, CI − 5 to 12%).

Soil CH_4 flux responses to biochar amendment depended on C/N ratio and pH of biochar (Table 1 and Figure 6B and C). Responses of soil CH_4 release to biochar amendment shifted from the highest positive response to biochar with the C/N ratio of below 50 (+57%, CI 43–79%) to the lowest negative response to biochar with the C/N ratio of above 300 (− 168%, CI − 214 to − 147%; Figure 6B). For upland soils acting as a sink of CH_4, negative responses of soil CH_4 uptake to biochar amendment consistently increased with the C/N ratio of biochar, but there was no significant difference in size of effect among them (Table 1). Acidic and slightly alkaline biochar (pH 7.0–8.5) significantly increased CH_4 release rates by 23 and 21% respectively, whereas highly alkaline biochar (pH 8.6–10 or > 10) led to a significant decrease by 99% or had no significant effect on CH_4 release (Figure 6C). For upland soils as a sink of CH_4, soil CH_4 uptake significantly and negatively responded to acid (− 124%, CI − 141 to − 87%) and extremely alkaline biochar (− 25%, CI − 79 to − 20%), as a contrast with positive response to slightly (+9%, CI 2–15%) and highly alkaline (+14%, CI 12–18%) biochar.

Responses of soil CH_4 release to biochar were significantly altered by pyrolysis temperature of biochar (Table 1 and Figure 6D), shifting from a significant positive response at pyrolysis temperatures below 400 °C (+25%, CI 18–44%) to the most negative response at temperatures above 600 °C (− 112%, CI − 144 to − 97%). In upland soils acting as a sink of CH_4, biochar generated under high pyrolysis temperatures (501–600 °C or > 600 °C) resulted in a significant decrease in soil CH_4

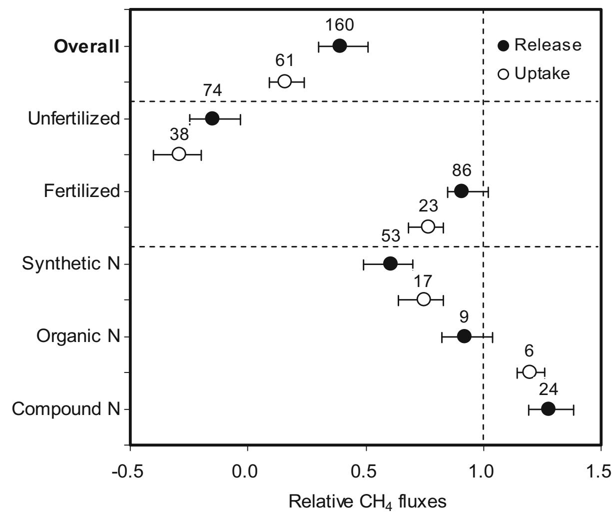

Figure 4. Soil CH_4 flux responses to biochar altered by fertilizer type and component sources, shown as relative fluxes (CH_4 release or uptake rate from controls are equal to one). Mean effect and 95% CIs are shown. Numerals indicate number of observations. 'Overall' indicates the integrated response with or without fertilizer application. The closed and open symbols represent original control soils acting as sources or sinks of CH_4, respectively. In organic and compound N applied soils, no available soils were sinks of CH_4.

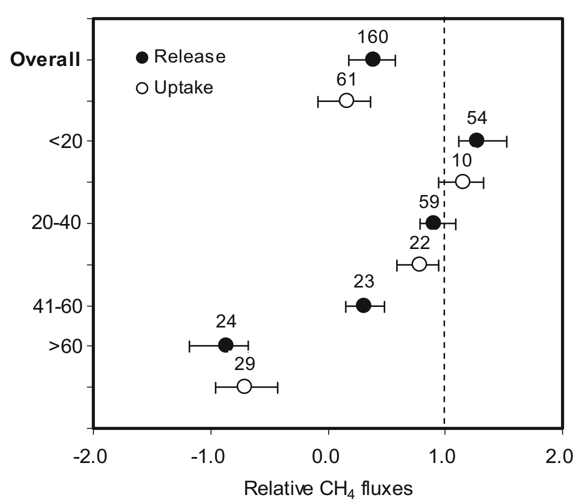

Figure 5. Response of soil CH_4 fluxes to biochar amendment affected by biochar applied rates (t ha^{-1}), shown as relative fluxes (CH_4 release or uptake rate from controls are equal to one). Mean effect and 95% CIs are shown. Numerals indicate number of observations. 'Overall' indicates the integrated effect across applied rates. The closed and open symbols represent original control soils acting as sources or sinks of CH_4, respectively. For the grouping categories of biochar applied rates < 20 t ha^{-1} or within 41–60 t ha^{-1}, no available soils were sinks of CH_4.

uptake, whereas no pronounced response to biochar was observed when pyrolysis temperatures fell to within 400–500 °C (− 8%, CI − 21 to 12%).

Robustness of Meta-analysis

In this study, removal of outlier datasets did not change the general results. After removing the outliers, the mean effect sizes of biochar treatments were − 57% for soil CH_4 release and − 86% for soil CH_4 uptake, similar to the effect sizes of − 61 and − 84% when including all datasets, respectively. After removing the data without any variances available, the mean effect sizes of biochar on soil CH_4 release and uptake were − 57 and − 78%, highly similar to − 61 and − 84% when averaged across all CH_4 flux measurements, respectively. Similarly, removing data without variance measures did not change the general results. However, a major weakness occurred when adopting this approach, because the size of datasets was considerably reduced from 222 to 169, and some category analyses could not be performed because of insufficient datasets available in the categories.

Discussion

Soil CH_4 Release Decreased by Biochar Amendment

Methane is mainly produced under anaerobic conditions where methanogenic archaea utilize soil C input by plants or soil organic materials amendment (for example, crop residue, biochar) as their ultimate source of organic substrates (Conrad 2007). Meanwhile, to different extents, soil pro-

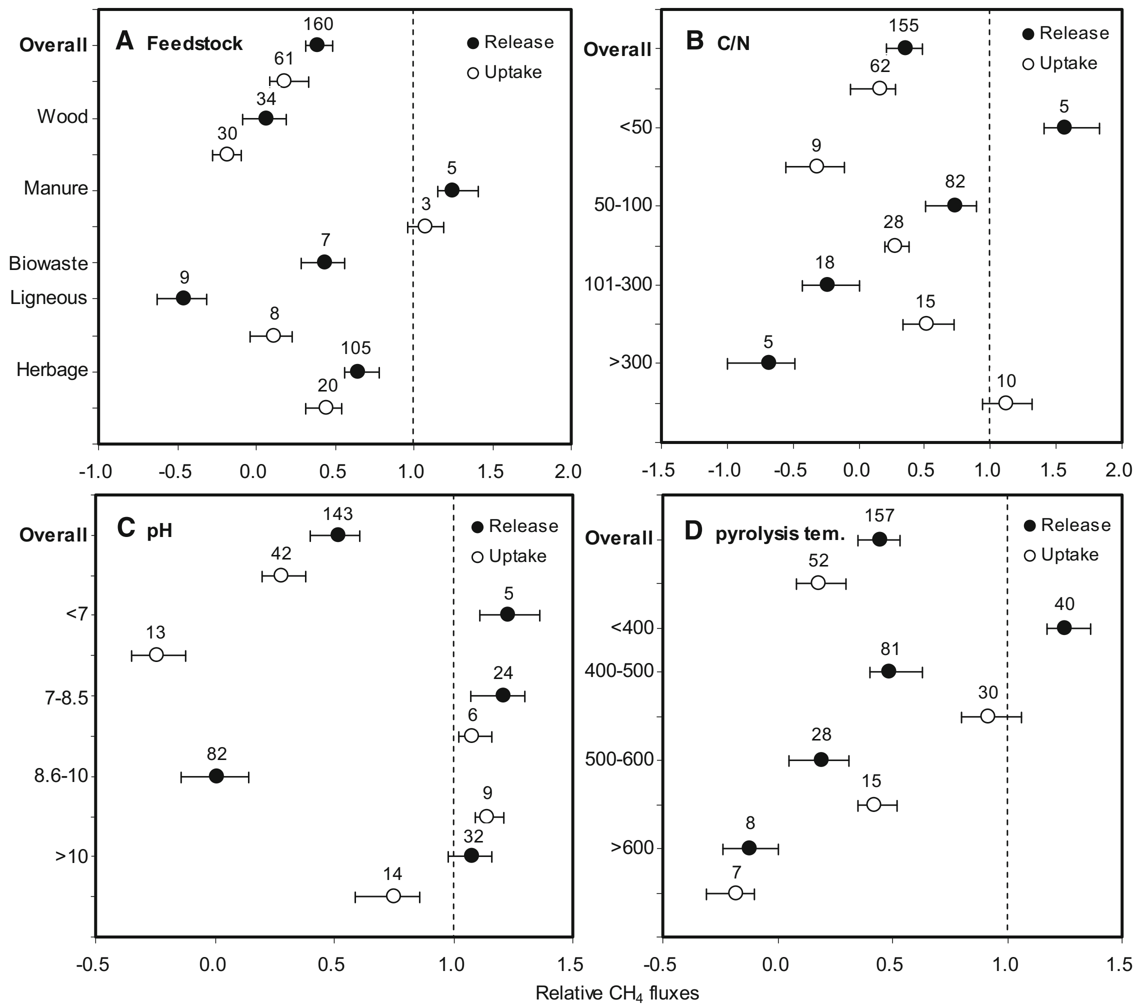

Figure 6. Response of soil CH_4 fluxes to biochar amendment dependent on biochar characteristics including feedstock (**a**), C/N ratio (**b**), pH (**c**) and pyrolysis temperature (**d**), shown as relative fluxes (CH_4 release or uptake rate from controls are equal to one). Mean effect and 95% CIs are shown. Numerals indicate number of observations. 'Overall' indicates the integrated effect within a given grouping category. The closed and open symbols represent original control soils acting as sources or sinks of CH_4, respectively. Among all grouping categories, no available soils amended with slight alkaline biochar (pH 7–8.5), biochar derived from biowaste or generated at low pyrolysis temperatures (< 400 °C) were sinks of CH_4 in this analysis.

duction of CH_4 is usually consumed by methanotrophs in soils. Therefore, soil CH_4 release rates depend on the combined performance of both methanogenic and methanotrophic communities. In this meta-analysis, soil CH_4 release rates were significantly decreased by biochar amendment both in paddy and upland soils (Figure 2), which is in accord with biochar-induced CH_4 mitigation potentials in some previous reports (Rondon and others 2005; Van Zwieten and others 2009). Several mechanisms have been underlined to explain biochar-induced decrease in soil CH_4 release. First, biochar typically has a large Brunauer–Emmett–Teller (BET) surface area, leading to a significant sorption capability of CH_4 in soils (Yaghoubi and others 2014). Second, the increase in oxygen supply due to biochar amendment can enhance soil aeration, promoting soil CH_4 oxidation in upland soils because microbial CH_4 oxidation in upland soils is mostly substrate-limited (Castro and others 1994). Third, the amendment of biochar with a high pH could decrease mcrA/pmoA ratios of soils because the size and/or structure of methanotrophic communities may be more sensitive to rising soil pH than that of methanogens (Feng and others 2012; Reddy and others 2014).

It is interesting that biochar-induced decreases in soil CH_4 release rates were larger in upland soils

(− 72%) than in paddy soils (− 12%) (Table 1 and Figure 2). In contrast to upland soils (Van Zwieten and others 2009), the aeration effect of biochar may be temporary and may disappear over time in paddy soils because of waterlogging. With this respect, soil CH_4 oxidization stimulated by biochar amendment would be weakened over time in paddy soils. This response difference could also be associated with more abundant Al^{3+}, Fe^{3+} and NH_4^+ in paddy soils relative to upland soils. Biochar can efficiently retain Al^{3+}, Fe^{3+} and NH_4^+ ions through adsorption (Liang and others 2006). These ions and CH_4 compete for oxidation by methanotrophs (Mosier and others 1991), and therefore, these ions retained by biochar can stimulate CH_4 emissions from paddy soils, which would partially negate mitigation effect of biochar on soil CH_4 releases from paddy soils.

Among different experimental methods, significant decreases in soil CH_4 release rates following biochar amendment were mainly contributed by controlled-environment studies (laboratory incubation or pot). Presumably, biochar-induced decrease in CH_4 release rates under controlled studies is most likely due to the relatively poor soil organic C (SOC) capacity for the soils enclosed in this category analysis as compared to in field conditions. On the other hand, field relative to controlled experimental conditions can augment the availability of microbial habitats and easy access of microbial food resources especially following biochar amendment, and thus stimulating soil methanogenic activities (Zackrisson and others 1996; Pietikäinen and others 2000). Nevertheless, the results of this meta-analysis suggested that incubation and pot experiments might have exaggerated the response of soil CH_4 release to biochar amendment.

The response of soil CH_4 release to biochar amendment depended on soil texture and pH (Figure 3). Biochar amendment in coarse and medium-textured soils significantly decreased CH_4 release rates, whereas biochar effects were positive in fine soils (Figure 3A). Relative to fine soils, the easier adequate mixture and larger contact area of biochar with soil particles in coarse- or medium-textured soils may greatly improve soil aeration and thus stimulate CH_4 oxidation. Biochar was effective at increasing soil CH_4 release rates in acid soils, although a significant negative response occurred in neutral and alkaline soils (Figure 3B). Several explanations may be given for increases in soil CH_4 release rates following biochar amendment in acid soils. First, biochar amendment to acid soils would neutralize the acid conditions, which makes it more adaptive for methanogenesis (Wang and others 1993; Yang and Chang 1997). Second, biochar in acid soils may trigger a more vigorous priming effect on decomposition of native organic matter, which would make more C substrates available for CH_4 production (Cross and Sohi 2011; Foereid and others 2011; Jones and others 2011). Eventually, biochar amendment can stimulate crop growth or biomass productivity in acid soils, providing more organic C substrate for CH_4 production (Liu and others 2013). Instead, biochar amended to neutral soils inhibited methanogenic activity, mainly due to enhanced soil pH (Liu and others 2011).

Biochar amendment alone significantly decreased soil CH_4 release rates, although this negative effect was largely weakened when biochar was amended in combination with N fertilizer (Figure 4). The results of this meta-analysis are in line with results of experimental studies, showing that biochar amended into N-limited soils had a high potential to decrease soil CH_4 release rates (Khan and others 2013; Mukherjee and others 2014). Biochar in N-poor soils would lead to further soil C immobilization and in turn greatly reduce the substrate for methanogenesis. Among N fertilizer types, biochar combined with compound N fertilizer tended to increase CH_4 release rates, in contrast to a negative response of soil CH_4 release when biochar amendment is combined with synthetic and organic N fertilizer. It is most likely that compound N fertilizer offered more balanced substrate for microbial methanogenesis in biochar-treated soils (Qin and others 2010). In general, responses of soil CH_4 release to biochar decreased with its applied rates (Figure 5). As evidence observed by Lehmann and others (2006), biochar amended at higher rates may suppress soil C mineralization as a high C/N ratio leading to low microbial N availability. Also, the decrease in CH_4 release rates at higher applied rates of biochar may be associated with specific biochar characteristics and their impacts on soil properties.

Biochar feedstock source and C/N ratio were important parameters influencing soil CH_4 release (Figure 6A and B). Soil CH_4 release rates were significantly increased by manure-derived biochar as compared with other feedstock sources in this meta-analysis (Figure 6A), which was presumably due to evidence that biochar was generated from manure materials with relatively low C/N ratios (grand mean of C/N ratio: 16). Amendment of biochar with a low C/N ratio has been found to result in an increase in bioavailable C sources (Baggs and others 2000; Cayuela and others 2010), and thus, there would be more soil C available for microbial processes such as methanogenesis (Major and others 2010; Lehmann and others 2011; Singh and others 2012). In contrast, the biochar derived from wood and ligneous materials with high C/N ratio had a negative effect on CH_4 release (Troy and others 2013; Mukherjee and others 2014). In addition, a negative response of CH_4 release occurred also for biowaste-derived biochar. It is most likely that biochar generated from biowaste (for example, municipal solid waste, sewage sludge) materials provided extremely limited substrate available for CH_4 production.

It is well documented that soil CH_4 release responses to biochar depended on biochar pH (Zhang and others 2010; Scheer and others 2011; Zheng and others 2012). In this meta-analysis, CH_4 release had a negative response to alkaline biochar (pH 8.6–10), in contrast to a positive response of CH_4 release to biochar with other pH values (< 7.0, 7–8.5 or > 10.0) (Figure 6C). As previously mentioned, the influence of biochar pH on soil processes largely relied on its pH buffering capacity for a given soil condition (Yuan and others 2011). The biochar with extreme pH values may be more beneficial to methanogenesis than methanotrophs, leading to an increase in the ratio of methanogenic to methanotrophic abundance in soils (Anders and others 2013; He and others 2017). On the other hand, biochar generated at low pyrolysis temperatures (< 400 °C) had a significant positive effect on CH_4 release, against significant negative responses to biochar at high pyrolysis temperatures (Figure 6D). In general, biochar created at high temperatures is more resistant to decomposition and thereby would better fulfill the C sequestration function (Novak and others 2010; Harvey and others 2012). Biochar created at low pyrolysis temperatures would be more suitable for improving soil nutrition balance and in turn promote soil microbial methanogenic activities, whereas biochar generated at high temperatures would generally lead to a material analogous to activated C (Ogawa and others 2006).

Soil CH_4 Uptake Decreased by Biochar Amendment

When averaged cross all upland soils acting as sinks of CH_4, soil CH_4 uptake was significantly decreased by biochar amendment. Decreased CH_4 uptake of upland soils under biochar application could be attributed to biochar compounds that inhibit the activity of methanotrophs (Spokas 2013). Besides, biochar can efficiently retain NH_4^+, Al^{3+} and Fe^{3+} through surface adsorption (Liang and others 2006). These ions compete with CH_4 for oxidation by methanotrophs and therefore inhibit soil CH_4 uptake but instead stimulate CH_4 emissions (Mosier and others 1991). This sorption generally increases with biochar applied rates, which is consistent with our results in upland soils, showing that the soil CH_4 uptake response to biochar decreased with rates of biochar application. Among different experimental methods, nevertheless, decreases in CH_4 uptake of upland soil following biochar amendment occurred only in controlled-environment studies. Presumably, relative to in upland field conditions, the methanotrophic oxidation of CH_4 in upland soils would be highly weakened in controlled laboratory or pot environments. Moreover, biochar effects in controlled experiments are generally amplified as compared to those in field studies, such as increased soil fertility, whereas microbial CH_4 oxidation in upland soils is mostly substrate-limited (Castro and others 1994).

Biochar amendment consistently decreased soil CH_4 uptake across soil textures. The consistent decrease in soil CH_4 uptake rates following biochar amendment across soil textures was contrary to some evidence obtained in individual studies, showing no significant effects of biochar on soil CH_4 uptake in upland soils (Wang and others 2012; Case and others 2014), but in agreement with the results of a meta-analysis proposed by Jeffery and others (2016). Biochar significantly decreased CH_4 uptake in both acid and neutral soils, whereas minor positive responses were observed in alkaline soils. As proposed by Rondon and others (2006), biochar amendment to acid or neutral soils relative to alkaline soils could greatly inhibit soil CH_4 oxidation, leading to a decrease in methanotrophs for enhanced soil aeration. The response of soil CH_4 uptake to biochar was quite dependent on N fertilizer and biochar application rates (Figures 4 and 5), suggesting that the interactive effect of biochar with N fertilizer and biochar application rates may contribute much to CH_4 uptake in biochar-treated upland soils. Spokas and Reicosky (2009) proposed that the effect of biochar on soil CH_4 fluxes may not only depend more on biochar characteristics, soil properties and habitat-specific climates, but applied rates.

Among biochar feedstock sources, negative effects of biochar on soil CH_4 uptake were the highest for biochar derived from ligneous materials. Given the limited datasets available and high level of variation within some grouping categories in this analysis, nevertheless, more studies are required to examine the response of CH_4 uptake to biochar as regulated by its feedstock sources. The soil CH_4 uptake response to biochar increased with the pH of biochar, shifting from a pronounced negative response to biochar with pH below 7.0 to a significant positive response to biochar with pH of 8.6–10.0. Presumably, the biochar with the high pH would inhibit soil CH_4 oxidation due to its induced low rate of soil C mineralization as a major energy source of methanotrophic activities (Crombie and others 2014). In addition, biochar created at high temperatures significantly decreased soil CH_4 uptake, suggesting that the high-temperature biochar-induced mitigation on soil CH_4 fluxes might not be attributed to improved soil CH_4 oxidation.

Merit of this Study and Future Concerns

This study first attempts to examine the biochar effect on soil CH_4 fluxes by distinguishing source and sink roles of soils. The results presented in this analysis further address limitations by previous reviews that did not investigate negative fluxes, preventing any conclusions to be drawn about soil CH_4 uptake. The evidence provided here demonstrates that biochar amendment benefits CH_4 mitigation for paddy soils, and a more beneficial effect can be achieved in upland soils acting as sources of CH_4. However, for upland soils acting as sinks of CH_4, a significant negative response of soil CH_4 uptake to biochar would instead potentially intensify global warming by increasing the source strength of atmospheric CH_4. Given that the global atmospheric CH_4 source strength of paddies was comparable to its sink strength of upland soils (Saunois and others 2016), biochar's potential to mitigate CH_4 emissions from paddies and uplands would be largely offset by its induced decreases in CH_4 uptake of upland soils. Overall, the results of this synthesis suggest that the role of biochar in soil CH_4 mitigation potential might have been exaggerated, particularly in fields where biochar is applied in combination with N fertilizer.

Although this meta-analysis provided an insight into soil CH_4 release and uptake responses to biochar amendment, the conclusions were only based on short-term experimental results. No studies were found in the literature extending more than two years and less than 75% of the studies have showed results over a whole growing season. Therefore, there is an urgent need for long-term studies to examine the effect of biochar on soil CH_4 fluxes with higher certainty.

Variance was heterogeneous among grouping categories as reflected in most figures, which was been expected for a meta-analysis enclosing data from a range of soil types, land-use types, and climatic regions. In some instances, the high level of variance (expressed by the 95% confidence) may be ascribed to the limited number of studies included within some certain categories. Indeed, further work is required to investigate whether the large variability originates from an inherent trait of a certain category reported or the limited number of datasets currently available within a given category.

In this meta-analysis, we could not fully take environmental and management factors into consideration, such as the auxiliary data on other soil key properties (for example, soil total organic or microbial C) due to the lack of relevant information in the literature, which may have interactive effects with biochar on soil methanogenesis or CH_4 oxidation processes. Therefore, to elucidate the sustainable effect of biochar on soil CH_4 fluxes, field experiments over a longer period across a wider range of environmental and management factors are needed in the future, instead of laboratory incubation or pot studies as included in the present quantitative analysis.

ACKNOWLEDGEMENTS

This work was supported by the Natural Science Foundation of China (41771268, 41771323), National Key Research and Development Program of China (2016YFD0201200), and Fundamental Research Funds for the Central Universities (KYTZ201404, KYZ201621).

Compliance with Ethical Standards

Conflict of interest The authors declare no conflict of interest.

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