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Biochar effects on methane emissions from soils: A meta-analysis

Running title: Meta-analysis of biochar effects on CH_4 flux

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Abstract

Methane (CH_4) emissions have increased by more than 150% since 1750, with agriculture being the major source. Further increases are predicted as permafrost regions start thawing, and rice and ruminant animal production expand. Biochar is posited to increase crop productivity while mitigating climate change by sequestering carbon in soils and by influencing greenhouse gas fluxes. There is a growing understanding of biochar effects on carbon dioxide and nitrous oxide fluxes from soil. However, little is known regarding the effects on net methane exchange, with single studies often reporting contradictory results. Here we aim to reconcile the disparate effects of biochar application to soil in agricultural systems on CH_4 fluxes into a single interpretive framework by quantitative meta-analysis.

This study shows that biochar has the potential to mitigate CH_4 emissions from soils, particularly from flooded (i.e. paddy) fields (Hedge's d = -0.87) and/or acidic soils (Hedge's d = -1.56) where periods of flooding are part of the management regime. Conversely, addition of biochar to soils that do not have periods of flooding (Hedge's d = 0.65), in particular when neutral or alkaline (Hedge's d = 1.17 and 0.44, respectively), may have the potential to decrease the CH_4 sink strength of those soils. Global methane fluxes are net positive as rice cultivation is a much larger source of CH_4 than the sink contribution of upland soils. Therefore, this meta-study reveals that biochar use may have the potential to reduce atmospheric CH_4 emissions from agricultural flooded soils on a global scale.

1. Introduction

Agriculture accounts for 10–12% of total global anthropogenic emissions of greenhouse gases (GHGs), which includes 50% of global methane (CH_4) emissions (Smith et al., 2007). Methane emissions have increased by 151% since 1750 (IPCC, 2007), and are currently increasing at a rate of 0.003 μmol mol^{-1} $year^{-1}$ (Butenhoff and Khalil, 2007; Bloom et al., 2010). Further increases are projected due to the growing demand for food, particularly animal protein, which could require *ca*. 70 million ha of additional land to fall under agricultural production (Alexandratos and Bruinsma, 2012).

Methane is primarily produced in water-logged anoxic soils by methanogenic archaea via methanogenesis (Conrad, 2007). Conversely, well-aerated upland soils are biological sinks for atmospheric CH_4 (Boone et al., 1993; Dunfield, 2007). Soil CH_4 uptake is driven by microbial oxidation of CH_4 by methanotrophs from groups including α- and γ-proteobacteria, a group of obligate aerobic bacteria some of which feed solely on CH_4 and others, along with genera such as *Methylocella* and *Methylocapsa*, that are facultive methanotrophs (Pratscher et al., 2011; Knief, 2015). Generally, both processes – methanogenesis and methanotropy – can occur simultaneously in micro-sites within the soil, or can be stratified with CH_4 production occurring in more highly anoxic depths, and CH_4 consumption occurring in overlaying oxic soil horizons. Here, the soil acts as a net source or sink depending on which is the overriding process (Hiltbrunner et al., 2012). However, these two processes can dynamically interact (Kammann et al., 2009) with CH_4 consumption functioning as a "biofilter" process that can ameliorate CH_4 emissions in various ecosystems, including rice paddies and landfill cover soils.

One of the main attractions underlying the biochar concept is the combination of soil carbon (C) sequestration with soil fertility (crop yield) increases (Glaser et al., 2001; Lehmann, 2007). Initial research efforts have focused on biochar's recalcitrance as a potential means to sequester C in soils (Lehmann et al., 2006; Nguyen and Lehmann, 2009; Gurwick et al., 2013) while concurrently increasing crop yields (Jeffery et al., 2011). It has also been shown to mitigate nitrous oxide (N_2O) emissions from agricultural soils (meta-analysis: Cayuela et al., 2014). The interactions between biochar and GHG fluxes such as carbon dioxide (CO_2) and N_2O, and the associated mechanisms, are becoming better understood (Cayuela et al., 2013; Maestrini et al., 2014; Cayuela et al., 2015; Obia et al., 2015; Sagrilo et al., 2015). However, there is still a paucity of information on CH_4 flux effects beyond the single study scale, which often report contradictory results.

Biochar has been shown to increase (Zhang et al., 2010; Spokas et al., 2011), decrease (Feng et al., 2012; Dong et al., 2013; Reddy et al., 2014), or have no significant effect (Kammann et al., 2012) on CH_4 emissions from soils. Mechanisms are usually only assumed or hypothesised and remain unclear. Meta-analysis is a useful tool for comparing results across studies to reveal common response patterns. It facilitates extrapolation of results and formulation of mechanistic hypotheses (e.g. within the same soil conditions; or with the same biochar types) and thus increases the robustness of extrapolations and predictions across systems.

The mechanisms by which biochar may affect soil CH_4 fluxes include sorption of CH_4 to biochar's surfaces (Yaghoubio et al., 2014), and soil aeration by biochar addition, which may increase diffusive CH_4 uptake (van Zwieten et al., 2010; Karhu et al., 2011), as microbial CH_4 oxidation in upland soils is mostly substrate-limited (Castro et al., 1994). However, in anoxic environments, the labile C pool of biochar may function as methanogenic substrate, promoting CH_4 production (Wang et al., 2012). Biochar has also been shown to promote methanotrophic CH_4 consumption at oxic/anoxic interfaces in anoxic environments, lowering CH_4 emissions via the "biofilter" function of CH_4 consumption (Feng et al., 2012; Reddy et al., 2014).

A recent work has also included meta-analysis of CH_4 emissions in response to biochar application as part of a wider analysis (Song et al., 2016). However, the method applied in their analysis does not allow the inclusion of negative fluxes (i.e. all CH_4 sinks) and thus was restricted in the conclusions that could be drawn. Here, we present the first comprehensive meta-analytical investigation of the effects of biochar application to soil in agricultural systems on CH_4 emissions drawing on studies with a global distribution.

2. Material and Methods

2.1. Data collection and categorisation

The keywords "biochar" AND "methane" OR "CH_4" were entered into the search engines of Scopus, Web of Science and Google Scholar to identify relevant studies for inclusion in the meta-analysis. This led to identification of 62 studies, to a cut-off date of 31st December 2014. Studies were vetted using inclusion criteria consisting of studies: (i) using a randomised design; (ii) using replicated samples per treatment; and (iii) containing a "treatment" and "control" such that the treatment was the same as the control in all aspects apart from the inclusion of biochar. Only cumulative net CH_4 fluxes were included. Where only daily or seasonal fluxes were reported, corresponding authors were contacted to ask for data on cumulative fluxes and means. When these data were provided the studies where included; otherwise they were excluded. Of the total studies, 42 met the inclusion criteria (Table S1), from which 189 pairwise comparisons were extracted.

Data were collected from tables presented in manuscripts where possible, or from figures using Plot Digitizer 2.6.6 (Huwaldt, 2015) or Web Plot Digitizer (Rohatgi, 2016), or from authors directly. Error bars were usually present in the form of standard errors; standard deviations were back calculated from these when necessary. When no measure of variance was available, corresponding authors were contacted to obtain such information. Categorical information concerning biochar, soil and environmental properties was also collected from manuscripts and recorded as auxiliary variables. These can be found in the full database which is available in supplementary information.

Auxiliary variables were grouped to facilitate cross-comparisons between studies using the same groupings as Cayuela et al. (2014). These variables related to *soil pH* grouped to *<6* and *6-8* and *>8* representing the optimum pH range for methanogenesis and methanotrophy; biochar *feedstock*, grouped as Manure - manures or manure-based materials from poultry, pig or cattle), Wood - oak, pine, willow, sycamore and unidentified wood mixtures, Herbaceous - greenwaste, bamboo, maize stover, straws; Biosolids – sewage sludge from water treatment plants and lignocellulosic wastes - including rice husk, nuts shells, paper mill waste; *pyrolysis temperature*, grouped as <450°C 450 – 600 and >600°C, $H:C_{org}$, grouped as <0.3 0.3-0.5 >0.5;, Brunauer, Emmett and Teller (BET) surface area (m^2 g^{-1}), grouped as *<100*, *100-500* and *>500*; water regime, water regime, grouped as Flooded (paddy soils and studies conducted under continuous waterlogged conditions), Cycles (paddy soils involving flooding-drying in which CH_4 emissions were measured during both the wet and dry periods) and Non-Flooded (studies were flooding was not part of the experimental setting); and N and Phosphorus (P) fertilization, grouped by rate for N, ≤120 kg N ha^{-1} and >120 kg N ha^{-1}, and as P or No P if P fertilizer was applied or not for P respectively.

2.2. Meta-analytical metric

Soils can function as both CH_4 sinks (negative values, uptake, consumption) and sources (positive values, emissions). The notation of the flux direction follows the convention by biogeochemists and takes the view from the atmosphere that gains or loses the gas in question. Since it is not possible to take a logarithm of a negative number, this precludes the use of the response ratio (calculated as the natural log of the experimental mean over the control mean) as a metric for comparison between studies, which is considered the preferred metric for ecological studies (Hedges et al., 1999). Here we utilise the standardised mean difference metric "Hedge's *d*" for analysis (Equation 1; Hedges and Olkin, 1985). This is a less biased indicator than "Hedge's *g*" (Equation 2; Hedges, 1981; Hedges and Olkin, 1985). Note that this is a different standardised mean difference metric to "Cohen's *d*" which was developed for behavioural science (Cohen, 1988); Hedge's *d* is less biased by small sample sizes (Hedges and Olkin, 1985) and was used as this was the case for most studies included in this meta-analysis.

Equation 1

$$d=\left(1-\frac{3}{4(n-2)-1}\right)g$$

Where n is the total sample size on which g is based, and g is Hedge's g as calculated by Equation 2

Equation 2

$$g = \frac{\bar{x}_1 - \bar{x}_2}{s}$$

Where $\bar{x}_1$ and $\bar{x}_2$ are the experimental and control means and s is the pooled standard deviation.

Here, experimental treatment refers to the treatment with biochar – controls are samples that are the same in all aspects, including any other amendment, without addition of biochar. A categorical random effects model was applied to *d*, with means weighted by the inverse of the variance. Confidence intervals (95%; CIs) were generated by bootstrapping (9999 iterations). To obtain a standardised mean effect size, the effect size was then divided by an estimate of the standard deviation of the effect sizes (Hedges and Olkin, 1985). Input data were arranged in Microsoft Excel 2010. Calculations were performed using Metawin Version 2 statistical software (Rosenberg et al., 2000).

The interpretation of the standardised mean effect size differs from the response ratio as it cannot be expressed as a percent change in response of an experimental treatment compared to a control. Rather, it is equivalent to a *Z*-score and as such represents the number of standard deviations that the standardised mean of the experimental treatment is from the standardised mean of the control. The effect of a response variable can be considered significant if the 95% CI does not intersect the standardised control mean (i.e. *Z*-value = 0). Groupings of auxiliary variables are considered significantly different if their 95% CIs do not overlap.

2.3. Interpretation of standardised mean effect size

There is no rigorously applied framework for interpretation of standardised means in terms of “effect sizes” because, unlike response ratios, they are probabilistic. That is, they describe the probability that a sample drawn from the control treatments would fall between the experimental mean and the control mean, assuming a normal distribution. By convention, a large effect is indicated by $d > 0.8$, a moderate effect by $d = 0.2 - 0.8$, and a small effect by $d = 0 - 0.2$ (Cohen, 1988; Gurevitch et al., 1992). However, it is generally acknowledged that these terms are relative and likely dependent on research area and methods (Hedges, 1981). A key point is that, using this metric, an effect size of (for example) 0.2 for a category does not equate to an effect size of 0.2 for categories in independent analyses presented in this paper, in absolute terms. Only categories within individual analyses, as differentiated by the horizontal dotted bars in Fig. 1 and 2, can be compared relatively (i.e. only within each category does an effect size of 0.4 equate to twice the size of 0.2; comparisons between figures are qualitative only). Further, small effect sizes (~0.2) may indicate significant changes in cumulative GHG fluxes, in absolute terms, particularly if effects persist over the long lifetime of biochar. Data are presented in two figures to allow use of different scale x-axis only and does not represent any fundamental difference in analyses.

Interpretation of effect sizes here is further confounded by the CH_4 sink/source flux direction in soils. A positive effect size implies a shift to the right on a scale going from strong net sink (i.e. negative flux values) to strong net source (i.e. positive flux values). However, it does not necessarily mean a change has occurred in the net sink/source status of the soil. Rather, it signifies that either the net sink strength has decreased, the soil has switched from sink to source, or that the net source strength has increased – and vice versa for negative effect sizes.

2.4. Control of biases

We tested the effects of publication bias using the Fail-safe *N* technique (Orwin, 1983; Rosenthal and Rosnow, 1991). A weakness with meta-analyses of experimental studies is that several experimental treatments are often compared to a single (identical) control in a published study. This artificially increases the number of replicate pairs and violates the assumption of independence that the effect size metric is based upon; the controls are necessarily counted repeatedly in pairwise control versus experimental treatment comparisons. Means of controlling for this bias (Borenstein et al., 2009; Aguilera et al., 2013) often show little effect (van Groenigen et al., 2006; Gattinger et al., 2012; Abalos et al., 2014; Skinner et al., 2014). Therefore, we here report results from the analysis on the level of single comparisons.

3. Results

Figure 1 shows the effect of biochar application to soils under different irrigation regimes. Biochar addition to *Flooded* soils (as part of their management practice) significantly increased in CH_4 sink strength / reduced source strength compared to *Flooded* soils without biochar application (Hedge's d = -0.87). Studies reporting biochar additions to *Non-Flooded* soils showed an overall moderate but significant decrease in the CH_4 sink strength / increase in source strength (Hedge's d = 0.65). Experiments in which irrigation was applied as *Cycles* of flooding and draining did not show a significant response to biochar application.

Biochar application to acidic soils (i.e. with a pH <*6*) resulted in the strongest effect size, causing a statistically significant increase in CH_4 sink strength / decrease in source strength following biochar application (Hedge's d = -1.56; Fig. 1). Conversely, addition of biochar to soils within the neutral pH range (i.e. *6-8*) showed a statistically significant decrease in CH_4 sink strength / increase in source strength (Hedge's d = 1.17). Application of biochar to soils with a pH greater than 8 did not show a statistically significant response to biochar application.

Biochar effects on CH_4 flux interact with N fertilizer rate (Fig. 1). Application of N fertilizers at rates less than 120 kg ha^{-1} caused a strong and statistically significant increase in CH_4 sink strength / decrease in source strength in the presence of biochar (Hedge's d = -3.1). Applications of N fertilizer at higher rates showed no interaction with biochar on soil CH_4 fluxes.

Biochars produced at high temperatures caused a statistically significant increase in CH_4 sink strength / reduction in source strength following application to soils (Hedge's d = -1.3; Fig. 1). Mid-temperature biochars (*450-600°C*) led to significant reductions in CH_4 sink strength / increased source strength when applied to soil (Hedge's *d* = 0.67).

In terms of interactions with feedstock source, biochar produced from *biosolids* led to a statistically significant increase in sink strength / reduction in source strength (Hedge's *d* = -6.03; Fig. 2). When produced from *Lignocellulosic waste*, biochar significantly decreased the CH_4 sink strength / increased the source strength (Hedge's *d* = 0.74). No other feedstock showed statistically significant effects on CH_4 fluxes.

No significant effects or differences between sub-groups were found for the category BET Surface Area; however, there was an apparent trend whereby increased BET surface area resulted in increasing sink strength / decreased source strength (Fig. 2).

4. Discussion

Using standardised mean differences as the meta-analysis metric precludes making firm conclusions in terms of changes in CH_4 sink/source functioning. A statistical approach based on measurements of net CH_4 fluxes alone does not enable differentiation between changes in methanogenesis or methanotrophy. However, it can identify the effect of biochar on the direction of net CH_4 fluxes (i.e. changes in overall sink/source strength). It also allows identification of the key management practices and soil and biochar properties which likely underlie the observed effects, and since the "usual" CH_4 flux direction in flooded wetland or aerated upland soils is known in general, the results provide first general insights into associated factors that need further investigation.

Application of biochar to soil produced a range of effects on CH_4 fluxes across studies, as expected. In most instances the "Grand Mean" (i.e. the mean response of all studies combined) was not significantly different to the control. This result is most likely due to contrasting responses (i.e. positive and negative effects on net CH_4 fluxes) cancelling each other out when studies assigned to all functional categories were combined. The data are unlikely to be significantly affected by publication bias, as studies finding either a positive or negative result are equally publishable, and most studies also investigated other factors such as N_2O fluxes and/or yield response. These have been shown to have a positive response to biochar application (Cayuela et al., 2014; Jeffery et al., 2011). As such, studies also investigating these metrics would have an increased chance of publication of the "associated" CH_4 flux results.

4.1. Irrigation management

Biochar addition to soils that were flooded as part of their management practice significantly increased CH_4 sink strength / reduced source strength compared to their controls. Methanogenesis is an exclusively anaerobic process (Thauer, 1998). Here (*Flooded*; Fig. 1), the change in CH_4 flux would likely equate to reduced net CH_4 emissions from flooded paddy soils, indicating that either the production decreased or methanotrophy in the rhizosphere increased through influencing the methanogenic/methanotrophic ratio of soils. Feng et al. (2012) reported that biochar decreased the ratio of methanogenic archaea to methanotrophic bacteria. In flooded soils, CH_4 consumption occurs at the aerated root interface where most CH_4 produced in the surrounding anoxic sediment usually enters the aerenchymatic root-shoot rice tissue, leaving the soil via this plant 'chimney'. Thus, increased CH_4 oxidation at the "biofilter" anoxic/oxic interface may explain the apparent CH_4 efflux mitigation potential of biochar application to flooded soils observed in our meta-analysis. Other studies in paddy soils (Liu et al., 2011; Singla et al., 2014) found no significant effects on methanogenic archaeal diversity between biochar treated and non-treated soils (but did not investigate CH_4 oxidizer communities).

Non-flooded (i.e. predominantly oxic) upland soils are an important sink for CH_4 and are considered to contribute to approximately 15% of global CH_4 oxidation (Powlson et al., 1997). Figure 1 suggests that biochar application may decrease net CH_4 oxidation by such soils. As intensively managed agricultural soils are relatively poor sinks of CH_4, it is likely that the decrease in net CH_4 efflux from *Flooded* soils more than counteracts any decrease in the net uptake from *Non-flooded* soils. Therefore, biochar use in rice agriculture may contribute to reducing the C footprint of rice production, which is usually worse than for example that of wheat production due to the CH_4 emission burden.

Experiments in which irrigation was applied as *Cycles* of flooding and draining did not show a significant response to biochar application. However, considerably fewer pairwise comparisons contributed to this category: 14 compared to 56 for *Flooded* and 85 for *Non-Flooded*. As such, there is reduced confidence in this result evidenced by the relatively large error bars (Fig. 1).

All studies included in this analysis were conducted in managed systems: either in the field or in controlled laboratory or greenhouse experiments. Currently, there is no work in the published literature that has investigated biochar effects when applied to natural wetlands, such as marshes, bogs and swamps, which can be significant sources of CH_4 emission (Bubier and Moore, 1994). This represents an unknown area of biochar research that may grow in importance as novel biochar applications are sought and potentially the biochar load of these systems increases due to biochar transport over time through waterways following erosion events (Jaffé et al., 2013).

4.2. Soil pH

Soil pH is one of the main environmental parameters that affects both methanogenesis and methanotrophy (Hanson and Hanson, 1996; Semrau et al., 2010). The optimum pH range of most methanogens ranges from 6 to 8 (Garcia et al., 2000), thereby overlapping with the optimum pH range for methanotrophy, which also extends to more acidic conditions (Le Mer and Roger, 2001; Semrau et al., 2010). Biochar generally has a higher pH than the soil to which it is applied, thereby providing a liming effect (Chidumayo, 1994; Yamato et al., 2006; Jeffery et al., 2011); the pairwise comparisons of this meta-analysis have an average pH of 6.2 for soil and 9.6 for biochar. As the optimum pH range for both methanogenesis and methanotrophy is similar, it may be expected that raising the soil pH to within the optimum range would affect both processes equally. However, we observed a significant increase in CH_4 sink strength / decrease in source strength for acidic soils (Fig. 1). A potential explanation is that the size and/or structure of methanotrophic communities may be more sensitive to rising soil pH than that of methanogens. Experiments quantifying, for example, the *mcrA*/*pmoA* ratios of soils are required to identify the cause underlying this observed effect.

Another possible explanation for the large CH_4 mitigating effect of biochar in acidic soils is related to Al^{3+} toxicity. Soils with a low pH are associated with increased Al^{3+} solubility, which is highly toxic for methanotrophic bacteria (Tamai et al., 2007). By increasing soil pH, biochar may reduce Al^{3+} release from cation exchange sites in the soil, thereby reducing toxicity levels for methanotrophs. A further analysis of initial soil pH effects on CH_4 fluxes, utilising a cut off at a pH of 5, the threshold above which Al^{3+} availability strongly decreases, provides further evidence for this explanation (Fig. S1). Biochar applied to soils with a pH <5 showed a significant increase in sink strength

/ reduction in source strength compared to soils with a pH >5. When biochar was applied to soils already above this threshold, no significant effect on CH_4 flux was observed. This hypothesis is in line with the literature on this topic (reviewed in Dunfield 2007; e.g. Sitaula & Bakken 2001) but more empirical studies are required to confirm or reject this hypothesised mechanism.

4.3. N Fertilizer

Figure 1 suggests that when biochar is applied with <120 t ha^{-1} N fertilizer, it can reduce CH_4 fluxes, while it has no effect when applied with >120 t ha^{-1} N. However, the effect of N fertilizer type and application rate on CH_4 flux is, in general, highly controversial. In soils where methanotroph N supply is not limiting to growth and activity, it is generally expected that the addition of NH_4^+-containing or delivering fertilizers will lead to decreased CH_4 oxidation due to competitive exclusion of CH_4 at binding sites by NH_4^+ (Bédard and Knowles, 1989; Sylvia et al., 2005). However, this effect is rate dependent; smaller amounts of N tend to stimulate CH_4 uptake while larger amounts tend to inhibit uptake into the soil (Aronson and Helilker, 2010). Despite this general rule, in severely N-limiting environments, the addition of an N source, even NH_4^+ which may also competitively inhibit CH_4 oxidation (Bedard and Knowles 1989; Gulledge et al., 1997), can lead to an increase in CH_4 oxidation due to an increase in methanotrophic biomass (Bodelier et al., 2000; Nazaries et al., 2013). The switch between stimulation and inhibition of CH_4 uptake has been reported to occur at between 100 kg N ha^{-1} (Aronson and Helilker, 2010) and 140 kg N ha^{-1} (Banger et al., 2012). As such, we set the threshold for our analysis to the mid-point between these studies - 120 kg N ha^{-1} (Fig. 1). This analysis shows that when N is applied above this threshold there is no significant difference between the experimental treatments (with biochar) and the controls (without biochar). When biochar is applied with levels of N below the threshold a significant difference is observed between the experimental treatments (with biochar) and the controls (without biochar) with increased sink strength/ reduced source strength being observed following biochar application with N fertilization rates below 120 k N ha^{-1}. The mechanism for this response pattern when biochar is applied with low N rates remains unclear and warrants further investigation

4.4. Pyrolysis temperature

Biochars produced at high temperatures caused a statistically significant increase in CH_4 sink strength / reduction in source strength following application to soils (Fig. 1). High temperature biochars are characterised by fewer labile compounds remaining on the surface of biochar particles, and so introduce less microbial substrate than lower temperature biochars when applied to soil (Brunn et al., 2011).

Reduced $H:C_{org}$ ratios in high temperature biochars indicate increased aromaticity, which is associated with the reducing effect of biochar on N_2O emissions (Cayuela et al., 2015). However, we did not find any relationship between $H:C_{org}$ and CH_4 fluxes from soil (Fig. S2).

Mid-temperature biochars (*450-600°C*) led to significant reductions in CH_4 sink strength / increased source strength when applied to soil (Fig. 1). The majority (73%) of the studies that used mid-temperature biochar were performed on non-flooded soils. This means that there is a confounding effect: it may be that the effect observed here is due to either biochar properties or soil water management - it is not possible to distinguish between the two with the analysis used for this study.

4.5. Feedstocks

In general, the feedstock from which biochar was produced did not lead to significantly different effect on CH_4 flux, with the exception of *biosolids* (Fig. 2). The effect size for biochar produced from biosolids is remarkably large (Hedges, 1981), as are the associated confidence intervals. This may be exacerbated by the low number of pairwise comparisons on which the statistic is based; all of the four pairwise comparisons were drawn from one study (Khan et al., 2013). The biochar used for this study was produced from sewage sludge (here grouped as *Biosolids*; according to Cayuela et al., 2014) and was applied to very acidic soil (i.e. pH = 4.02). Possible mechanisms, as discussed above, include potential changes in the size and/or structure of methanotrophic communities, or potentially reduced Al^{3+} toxicity effects. In addition, the effect may also be partly due to the high sulphur content of this feedstock (5.3% dry weight). This hypothesis is consistent with previous results that showed decreased CH_4 emissions when ammonium sulphate was used as a fertilizer compared to urea (Bufogle et al., 1998).

Biochar produced from *Lignocellulosic waste* led to a significantly decreased CH_4 sink strength / increased source strength. The mechanism underlying this effect remains unclear and warrants further research.

4.6. BET Surface Area

Biochar production temperature and the Brunauer, Emmett and Teller (BET) surface area of biochars have been shown to be positively correlated (Ronsse et al., 2013; Kambo and Dutta, 2015). This suggests that adsorption of CH_4 to the surface of biochars (Sadasivam and Reddy, 2014) may also be responsible for the reduced flux in the high temperature biochars (Fig. 1). However, this characteristic is often not reported in biochar studies, which hinders investigation of this potential mechanism. It appears that there is a trend whereby increased BET surface area results in decreased CH_4 flux (Fig. 2). However, the data are highly variable in the highest category (*>500*) with little confidence in the mean value owing to the low number of pairwise comparisons on which this statistic is based (n = 3). More studies using high surface area biochars, or systematically varying BET, are needed to investigate the importance of CH_4 or inhibitory N adsorption onto biochar as a mechanism underlying observed reductions in CH_4 fluxes.

5. Conclusions

Evidence presented in this study shows that biochar does have the potential to mitigate CH_4 emissions from soil, particularly from paddy fields and/or acidic soils that use periods of flooding as part of their management regime. However, addition of biochar to neutral or alkaline soils that do not have periods of flooding, may have the potential to decrease the CH_4 sink strength of those soils. These results indicate that soil and biochar properties, as well as management conditions, must be considered to maximise biochar's potential to mitigate CH_4 emissions and minimise trade-offs.

This meta-analysis highlights the importance of reporting key functional characteristics of biochar properties. Biochar pH has been shown to be highly pertinent for predicting response of some ecosystem functions to biochar application, in both this current study and previous studies (Jeffery et al., 2011; Sagrilo et al., 2015). Other functional characteristics (or proxies thereof) such as the molar $H:C_{org}$ ratio are becoming more recognised as effective predictors (Cayuela et al., 2015). Here we show that BET surface area may be an important functional characteristic in terms or predicting CH_4 flux mitigation potential of biochar. However, insufficient numbers of experiments have reported the characteristic to draw firm conclusions. It is vital that biochar researchers characterise and report functional characteristics of their biochars wherever possible.

Finally, it is apparent that trade-offs are inevitable and clear goals are necessary before effective advice can be offered to land managers and policy makers (Jeffery et al., 2015). For example, low temperature, slow pyrolysis maximises biochar production (Sohi et al., 2010) and thereby also C sequestration potential. However, evidence presented in this study shows that high temperature biochars are more effective at mitigating CH_4 emissions (the same applies for N_2O, Cayuela et al., 2015). Which one has the greatest potential to mitigate climate change thus remains to be determined and will require life cycle assessment approaches. However, market forces are likely to make the former more attractive until the full environmental costs of production are included as part of agricultural products.

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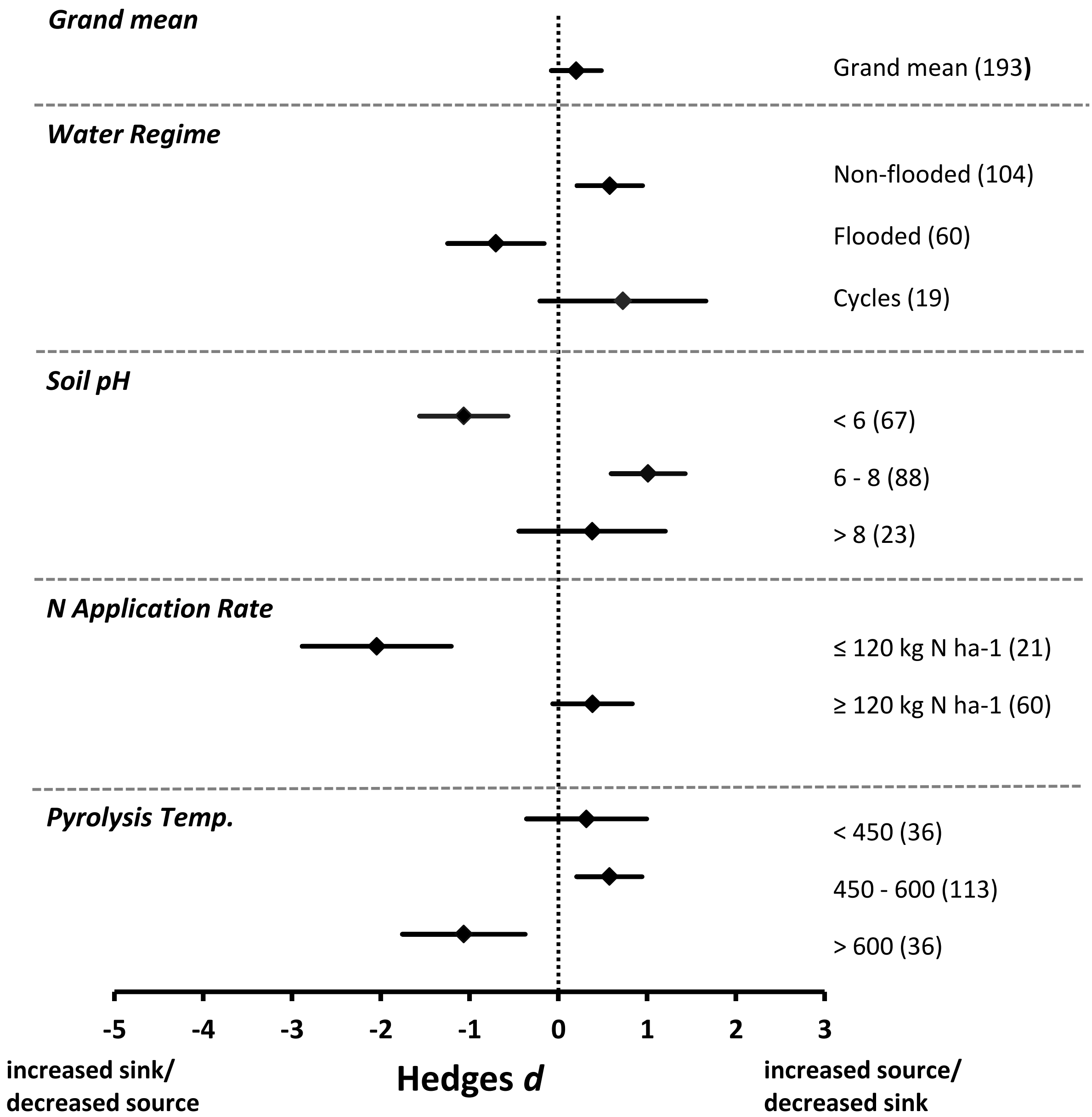
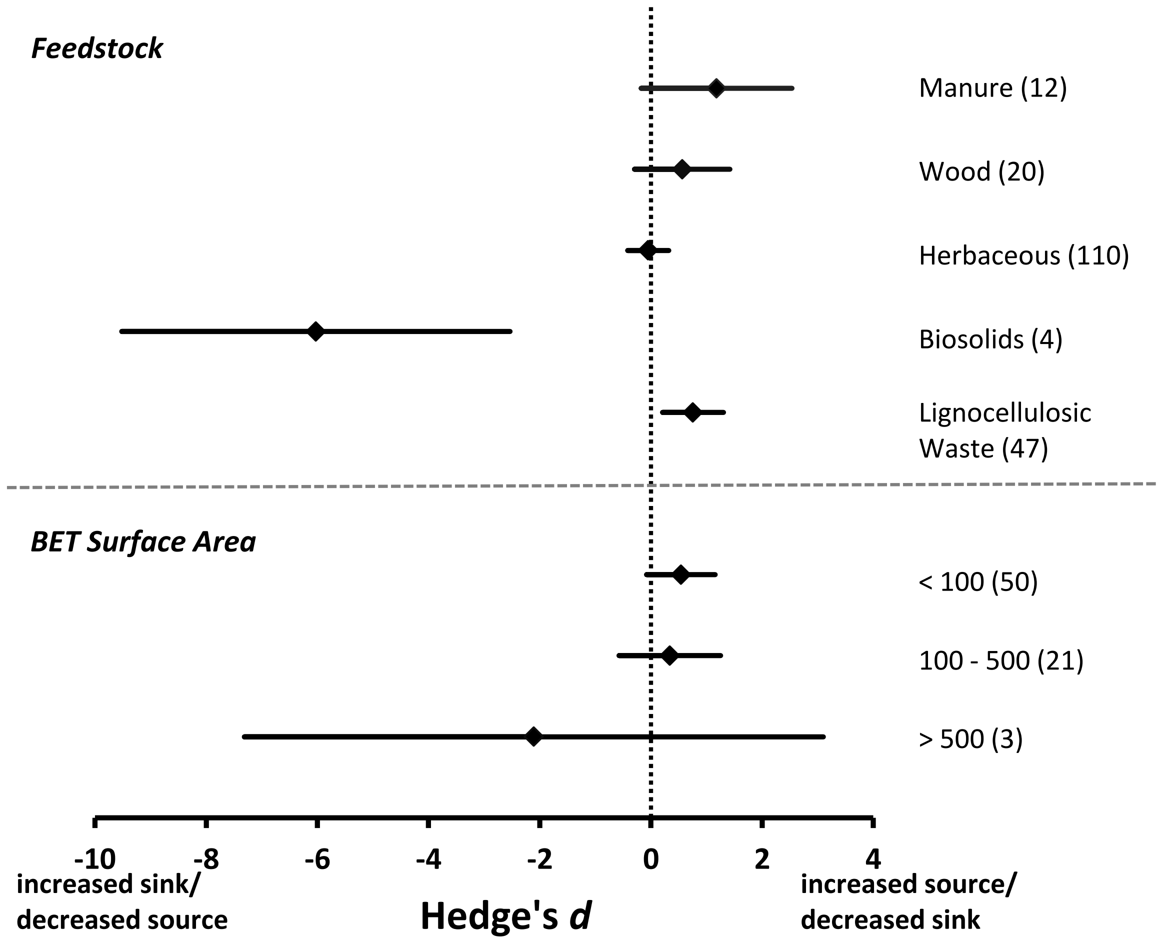
Figure 1. A forest plot of Hedge's *d* calculated from published literature grouped by experimental water regime, soil pH pre-biochar amendment, N fertilizer application rate and biochar pyrolysis temperature. Points show means, bars show 95% confidence intervals. The numbers in parentheses indicate the number of pairwise comparisons on which the statistic is based. (For an explanation of the Hedge's *d* metric see text).

Figure 2. A forest plot of Hedge's *d* calculated from published literature grouped by biochar feedstock type and BET (Brunauer, Emmett and Teller) surface area. Points show means, bars show 95% confidence intervals. The numbers in parentheses indicate the number of pairwise comparisons on which the statistic is based. (For an explanation of the Hedge's *d* metric see text).



Grand mean
Grand mean (193)
Water Regime
Non-flooded (104)
Flooded (60)
Cycles (19)
Soil pH
< 6 (67)
6 - 8 (88)
> 8 (23)
N Application Rate
≤ 120 kg N ha-1 (21)
≥ 120 kg N ha-1 (60)
Pyrolysis Temp.
< 450 (36)
450 - 600 (113)
> 600 (36)
-5 -4 -3 -2 -1 0 1 2 3
increased sink/
decreased source
Hedges d
increased source/
decreased sink



Feedstock
Manure (12)
Wood (20)
Herbaceous (110)
Biosolids (4)
Lignocellulosic
Waste (47)
BET Surface Area
< 100 (50)
100 - 500 (21)
> 500 (3)
-10
-8
-6
-4
-2
0
2
4
increased sink/
decreased source
Hedge's d
increased source/
decreased sink

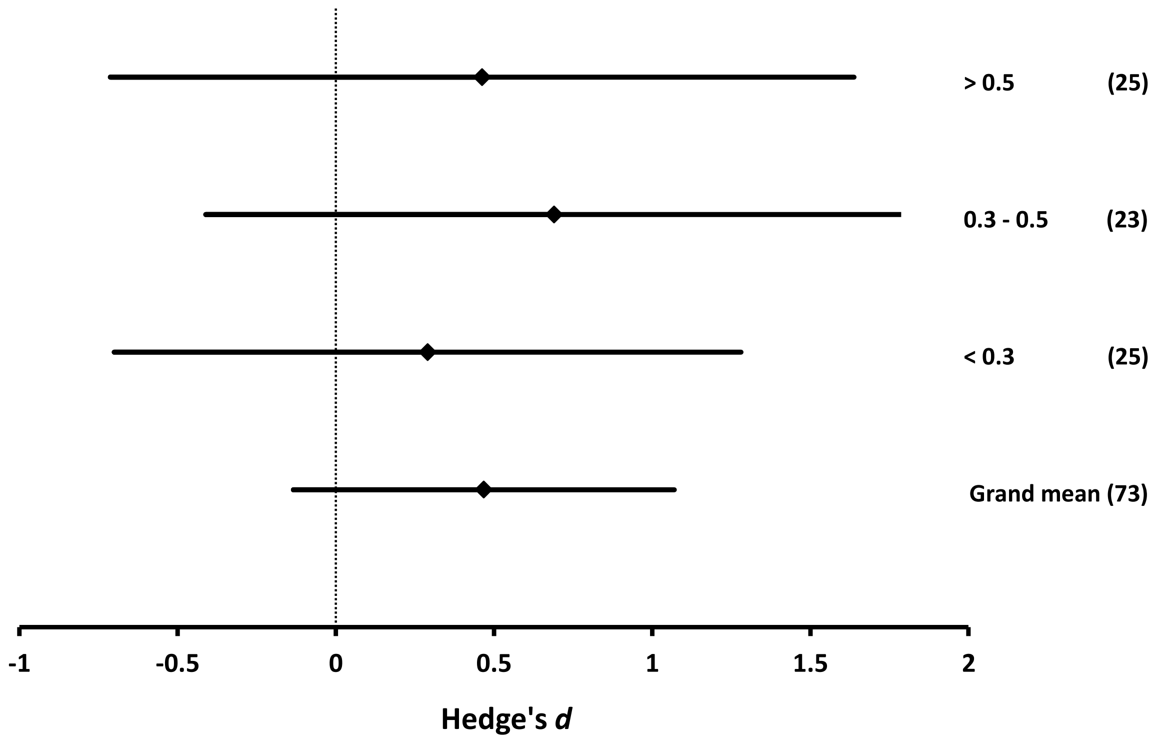

> 0.5 (25)
0.3 - 0.5 (23)
< 0.3 (25)
Grand mean (73)
-1
-0.5
0
0.5
1
1.5
2
Hedge's *d*

Figure S1. A forest plot of Hedge's d calculated from published literature grouped by pre-biochar amendment soil pH. The pH 5 threshold is applied to investigate the potential effects of aluminium bioavailability/toxicity. Points show means, bars show 95% confidence intervals. The numbers in parentheses indicate the number of pairwise comparisons on which the statistic is based. (For an explanation of the Hedge's *d* metric see text).

Figure S2. A forest plot of Hedge's *d* calculated from published literature grouped by H:C_{org}. Points show means, bars show 95% confidence intervals. The numbers in parentheses indicate the number of pairwise comparisons on which the statistic is based. (For an explanation of the Hedge's *d* metric see text).

Table S1. A list of the studies included in the meta-analysis database.

Reference	*Country*	*Soil pH*	*Water regime*	*BET surface area*	*Pyrolysis temperature*	*H:Corg*	*Biochar feedstock*
Castaldi et al., 2011	Italy	<6	Non-flooded	-	450-600	-	Wood
Dong et al., 2013	China	<6	Flooded	<100	>600	0.3-0.5, >0.5	Herbaceous
Feng et al., 2012	China	6-8	Flooded	-	<450, 450-600	-	Herbaceous
Fungo et al., 2014	Kenya	6-8	Non-flooded	-	<450, 450-600	0.3-0.5, >0.5	Herbaceous
Khan et al., 2013	China	<6	Flooded	<100	450-600	-	Biosolids
Liu et al., 2011	China	<6	Flooded	-	>600	0.3-0.5, >0.5	Herbaceous
Scheer et al., 2011	Australia	<6	Non-flooded	-	450-600	>0.5	Herbaceous
Singla and Inubushi, 2014	Japan	<6	Flooded	-	<450	-	Manure
Spokas et al., 2009	USA	6-8	Flooded	<100	450-600	0.3-0.5	Lignocellulosic
Wang et al., 2012	China	6-8	Flooded	-	450-600	-	Lignocellulosic
Wu et al., 2013	Canada	<6	Non-flooded	-	450-600	0.3-0.5	Herbaceous
Xie et al., 2013	China	6-8	Cycles	-	<450	-	Herbaceous
Zhang et al., 2012	China	6-8	Cycles	<100	450-600	-	Herbaceous
Zhang et al., 2012	China	>8	Non-flooded	<100	450-600	-	Herbaceous
Zhang et al., 2010	China	6-8	Flooded	<100	450-600	-	Herbaceous
Zheng et al., 2012	USA	6-8, >8	Non-flooded	100-500	450-600	<0.3	Lignocellulosic
Liu et al., 2014	China	<6	Flooded	-	450-600	-	Herbaceous
Pandey et al., 2014	Vietnam	-	Cycles	-	-	-	Herbaceous
Schimmelpfennig et al., 2014	Germany	<6	Non-flooded	>500	>600	<0.3	Herbaceous
Shen et al., 2014	China	<6	Flooded	-	-	-	Herbaceous
Singla et al., 2014	Japan	6-8	Flooded	-	<450	>0.5	Manure
Zhao et al., 2014	China	6-8	Flooded	-	450-600	-	Herbaceous
Zhang et al., 2014	Canada	6-8	Non-flooded	-	>600	-	Lignocellulosic

Zhang et al., 2013	China	6-8	Cycles	<100	450-600	-	Herbaceous
Jia et al., 2012	China	<6	Non-flooded	-	<450	-	Herbaceous
Ali et al., 2013	Bangladesh	6-8	Cycles	-	450-600	-	Lignocellulosic
Spokas et al., 2013	USA	6-8	Non-flooded	<100	450-600	<0.3, 0.3-0.5	Lignocellulosic
Angst et al., 2014	USA	6-8	Non-flooded	<100	450-600	>0.5	Lignocellulosic
Case et al., 2014	UK	6-8	Non-flooded	-	<450	-	Lignocellulosic
Li et al., 2013	China	<6, >8	-	<100	<450, 450-600	-	Herbaceous
Ly et al., 2014	Cambodia	<6	Flooded	-	450-600	-	Herbaceous
Stewart et al., 2013	USA	>8	Non-flooded	100-500	450-600	<0.3	Lignocellulosic
Watanabe et al., 2014	Japan	-	Non-flooded	-	>600	-	Herbaceous
Karhu et al., 2011	Finland	-	Non-flooded	<100	<450	-	Lignocellulosic
Troy et al., 2013	Ireland	6-8	Non-flooded	-	>600	-	Manure
Mukherjee et al., 2014	USA	6-8	Non-flooded	100-500	>600	-	Wood
Thomazini et al., 2015	USA	<6, 6-8	Non-flooded	<100	450-600	-	Wood
Vu et al., 2015	Vietnam	<6	Cycles	-	-	-	Herbaceous
Zhang et al., 2015	China	6-8	Cycles	<100	450-600	-	Herbaceous
Li et al., 2015	China	<6	Non-flooded	<100	<450	-	Herbaceous
Lin et al., 2015	China	>8	Non-flooded	-	<450	-	Herbaceous
Yoo et al., 2015	Korea	<6, 6-8	Flooded	-	<450, >600	0.3-0.5, >0.5	Herbaceous
Yu et al., 2013	China	<6	Non-flooded, Flooded	-	-	>0.5	Manure

Supplementary Table S2. Between-group heterogeneity (Q_b), within-group heterogeneity (Q_w) and total heterogeneity (Q_t).

	Q_b	Q_w	Q_t
Water regime	16.55***	342.32***	358.88***
Soil pH	40.92***	324.99***	365.92***
N application rate	27.76***	117.67**	145.44***
Pyrolysis temperature	17.98***	349.95***	367.93***
Feedstock	41.46***	348.31***	389.77***
BET Surface Area	5.41	148.12***	153.54***
Soil pH - cut off at pH 5	6.45*	350.41***	356.86***
$H:C_{org}$ molar ratio	0.31	180.76***	181.07***
Soil texture	7.76*	238.73***	246.49***

*$p<0.05$; **$p<0.01$; ***$p<0.001$