



Review

Biochar, soil and land-use interactions that reduce nitrate leaching and N₂O emissions: A meta-analysis



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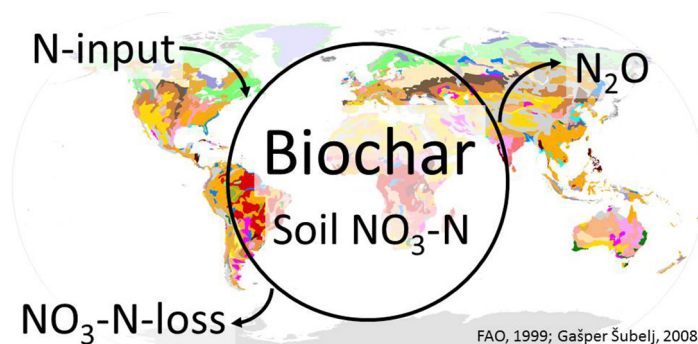
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HIGHLIGHTS

- N₂O emissions were reduced by 38% with biochar.
- Soil NO₃⁻ concentrations remained unaffected.
- NO₃⁻ leaching was reduced by 13% with biochar.
- Biochar strongly reduced N₂O-emission in paddy and sandy soils.

GRAPHICAL ABSTRACT



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ABSTRACT

Biochar can reduce both nitrous oxide (N₂O) emissions and nitrate (NO₃⁻) leaching, but refining biochar's use for estimating these types of losses remains elusive. For example, biochar properties such as ash content and labile organic compounds may induce transient effects that alter N-based losses. Thus, the aim of this meta-analysis was to assess interactions between biochar-induced effects on N₂O emissions and NO₃⁻ retention, regarding the duration of experiments as well as soil and land use properties. Data were compiled from 88 peer-reviewed publications resulting in 608 observations up to May 2016 and corresponding response ratios were used to perform a random effects meta-analysis, testing biochar's impact on cumulative N₂O emissions, soil NO₃⁻ concentrations and leaching in temperate, semi-arid, sub-tropical, and tropical climate. The overall N₂O emissions reduction was 38%, but N₂O emission reductions tended to be negligible after one year. Overall, soil NO₃⁻ concentrations remained unaffected while NO₃⁻ leaching was reduced by 13% with biochar; greater leaching reductions (>26%) occurred over longer experimental times (i.e. >30 days). Biochar had the strongest N₂O-emission reducing effect in paddy soils (Anthrosols) and sandy soils (Arenosols). The use of biochar reduced

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Nitrous oxide
Nitrogen
Soil organic carbon

both N₂O emissions and NO₃⁻ leaching in arable farming and horticulture, but it did not affect these losses in grasslands and perennial crops. In conclusion, the time-dependent impact on N₂O emissions and NO₃⁻ leaching is a crucial factor that needs to be considered in order to develop and test resilient and sustainable biochar-based N loss mitigation strategies. Our results provide a valuable starting point for future biochar-based N loss mitigation studies.

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1. Introduction

Agriculture accounts for ~60% of global anthropogenic N₂O emissions, largely due to organic and mineral nitrogen (N) fertilizer use and the extended use of legumes either as crops (soy, pea, bean or groundnut) or as green cover (Davidson, 2009; IPCC, 2013; Kammann et al., 2017; Smith et al., 2008). In addition to fertilizer-induced N₂O emissions, excessive N fertilization or inadequate timing of N application not fitting plant demand also leads to N leaching that affects ground and surface water quality, reduces N use efficiency (Ding et al., 2010), and subsequently elevates indirect N₂O emissions (Cooper et al., 2017; Minghua et al., 2017; Tian et al., 2017) e.g. from landscape-draining waterways (Turner et al., 2015).

Soil N₂O emissions and nitrate (NO₃⁻) losses are mainly a result of microbial activities affecting inorganic soil N concentrations via nitrification and denitrification processes, and by abiotic processes (Barnard et al., 2005; Bateman and Baggs, 2005; Cayuela et al., 2014; IPCC, 2006). Nitrification is the transformation of ammonium (NH₄⁺) to NO₃⁻ via nitrite (NO₂⁻), with N₂O being a by-product. Denitrification reduces NO₃⁻ or NO₂⁻ to NO, N₂O, and N₂ (Barnard et al., 2005; Cayuela et al., 2014; Harter et al., 2014; IPCC, 2006; Kammann et al., 2017). Efficiency and productivity of nitrification is mainly affected by availability of N and oxygen while availability of biodegradable organic matter and lack of oxygen govern efficiency and productivity of denitrification (Barnard et al., 2005; Bateman and Baggs, 2005; Linn and Doran, 1984; Granli and Bockman, 1994). Microbes performing nitrification prefer slightly acidic to alkaline soil pH, while soil denitrification is at optimum between pH 4 to 8 (Ahn, 2006; Antoniou et al., 1990; Barnard et al., 2005). Availability of easily biodegradable organic matter stimulates denitrification as it provides energy to maintain microbial metabolism and electrons required to reduce NO₃⁻ (Ahn, 2006; Barnard et al., 2005). Thus, physico-chemical soil properties regulate and organic matter amendments stimulate N₂O emissions.

Recently, biochar has been proposed as an organic carbon (C) soil amendment for reducing leaching of soil compounds (Abdelrahman et al., 2018; O'Connor et al., 2018a) and for improving soil quality (Crane-Droesch et al., 2013; Liu et al., 2013; Mehmood et al., 2017). Biochar may add both a small mineralizable and a more recalcitrant, less mineralizable C fraction to soils (Wang et al., 2016); additionally, biochar has also been shown to retain NO₃⁻ within its pores (Kammann et al., 2015; Haider et al., 2016, 2017; Hagemann et al., 2017a; Sumaraj and Padhye, 2017). Therefore, applying biochar to soils may affect conditions that control nitrification, denitrification (Cayuela et al., 2014; Kammann et al., 2017; Liu et al., 2018), and other N transformation and loss pathways. In order to evaluate biochars' overall potential and magnitude for reducing N losses across application locations, agricultural systems, application rates, and time, meta-analyses have been found to be a useful tool (Borenstein et al., 2009).

The first meta-analysis studying biochar impact on soil N₂O emissions showed a mean reduction of 54% (Cayuela et al., 2014); further meta-analyses, particularly those including field studies, have shown lower N₂O reductions ranging between 12 and 32% (Cayuela et al., 2015; Liu et al., 2018; Verhoeven et al., 2017). The meta-analysis published by Liu et al. (2018) was the first study to present biochar impacts on N pools, N fluxes, and the N cycle, but interactions between biochar use and experimental duration, soil types, and land use are still scarcely understood or exclusively assessed for C cycling (Wang et al., 2016; Verhoeven et al., 2017; Duarte-Guardia et al., 2018). Thus, the current meta-analysis differs from previous biochar meta-analyses, because it assesses the impact of biochar on N₂O emissions, NO₃⁻ leaching and final NO₃⁻ concentration in soil based on: (i) experimental duration that affects the C and N cycle (Hagemann et al., 2017b; Wang et al., 2016), (ii) relevance of soil and land use types to provide a basis for spatial (global) assessments (Duarte-Guardia et al., 2018; Werner et al., 2018), and (iii) impact of agricultural practices, such as vegetation type and fertilizer use.

2. Material and methods

2.1. Data compilation

A comprehensive survey of literature published between January 1, 2010 and May 31, 2016 was conducted, compiling 608 observations from 88 peer-reviewed publications accessed on the ISI Web of Knowledge. By using the term “biochar” in the “topic” field, 3328 publications appeared, but the number was reduced to 88 publications by abstract and full publication screenings (Table S1). Studies were scrutinized using the following inclusion/exclusion quality criteria: They: (i) were conducted in soil (e.g., horticultural substrates were excluded); (ii) included a minimum of three replicates per treatment; (iii) followed a randomized design; (iv) contained a “treatment” and a “control” such that the treatment was the same as the control in all aspects except for the inclusion of biochar; and (v) reported cumulative net N₂O emissions, cumulative NO₃⁻ leached and/or final NO₃⁻ concentrations in soil. Data (i.e. mean values, standard deviation, standard error, number of replicates) on N₂O fluxes, NO₃⁻ concentration in soil after experiment, and NO₃⁻ leaching were collected from tables, from figures by using WebPlot Digitizer software (www.automeris.io/WebPlotDigitizer/), or from contacting the authors directly. The final dataset consists of 608 observations, with 120 observations for nitrate leaching, 146 observations for nitrate concentration in soil after the experiment, and 435 observations for cumulative N₂O emissions (Table S2). Factors controlling N₂O emissions, NO₃⁻ concentration in soil after the experiments, and cumulative NO₃⁻ leaching were also collected from the publications. Once the dataset was completed, it was subjected to a strict quality check and each observation and related predictor variable was checked by at least two researchers independently. When there was a disagreement between the extracted values, the respective study was checked by an additional third researcher and a final correction was reached. Data were then grouped by study length, biochar properties, soil properties, N fertilization, and land use. Grouping was performed in accordance to previous classifications used by Jeffery et al. (2011) and Cayuela et al. (2014) (Table S3), considering a minimum number of observations per group to process reliable data (Table S4).

2.2. Meta-analysis

The effect of biochar treatment on N₂O emissions, NO₃⁻ concentration, and NO₃⁻ leaching was estimated via meta-analysis. As a standardized metric of the effect size, the natural log response ratio (RR) was computed for each experiment (Hedges et al., 1999):

$$RR = \ln\left(\frac{X_{trt}}{X_{ctr}}\right),$$

which is the ratio between the treatment mean (X_{trt}) and the control mean (X_{ctr}). In the case of N₂O emissions, RR is defined as the ratio between the cumulative N₂O emissions from the biochar treated soil and the N₂O emissions from the non-treated soil in each study. An RR with zero would mean no effect, while a negative or positive RR value would mean a reduction or increase in N₂O emissions through biochar treatment, respectively. The logarithm ensures better statistical properties of the effect size distribution and equal influence of nominator and denominator on the metric. The RR was expressed as a percentage change relative to zero. The variance of RR is given as (Hedges et al., 1999):

$$var(RR) = \frac{SD_{trt}^2}{N_{trt}X_{trt}^2} + \frac{SD_{ctr}^2}{N_{ctr}X_{ctr}^2},$$

where SD_{trt}^2 and SD_{ctr}^2 are the standard deviation and N_{trt} and N_{ctr} are the sample sizes of the treatment or control of the experiment. The parameters for calculating RR and $var(RR)$ were extracted from the studies or

recalculated when necessary. Funnel plots were used to detect biases in the traits included in the meta-analyses. The funnel plots were symmetric for N₂O emissions, NO₃⁻ concentration, and NO₃⁻ leaching data sets, which indicates absence of publication biases.

The combined effect size over all available studies was estimated with a random effects model. The random effects model was chosen because we did not assume that the underlying true effect size is homogeneous over all included studies due to study conditions and environmental influences, and we further wanted to make generalizations beyond the observed studies (Hedges and Vevea, 1998). The random effects model was estimated with the DerSimonian-Laird estimator (DerSimonian and Laird, 2015). Each study was weighted by the inverse of its sampling error variance (inverse-variance-weighting), which ensures that studies with very small sample sizes do not have a severe influence on the estimates. The overall effect was estimated for cumulative N₂O emissions, final NO₃⁻ concentration, and cumulative NO₃⁻ leaching. For assessing the heterogeneity of the meta-analysis, the I² index was used. The I² index indicates the percentage of the total variability among the effect sizes that can be explained by the between-studies heterogeneity (Huedo-Medina et al., 2006). Furthermore, we explored possible factors and relations influencing the overall effect sizes including study length, biochar properties, soil properties, N fertilization, and land use. These factors were grouped in accordance to previous classifications by Cayuela et al. (2014) and Jeffery et al. (2011) (Tables S3 & S4). Categories with less than two samples were removed from analysis. The subgroup analysis was conducted with a categorical random effects model and summarized in forest plots. All estimates were reported along with 95% confidence intervals. Estimates and confidence intervals were calculated from bootstrapping the random effects models with 1000 bootstrap intervals. Positive publication bias was tested with the Failed Safe N-test that takes into account the tendency of journals to only publish significant results. A fail-safe number was calculated using the Rosenberg method, indicating the number of non-significant or missing studies that one would need to add to the meta-analysis data set to reduce the observed, overall statistically significant results (Rosenberg, 2007).

3. Results

3.1. Percent overall changes and their change over time

Results showed a significant reduction of overall N₂O emissions by 38% ($P < 0.05$; Fig. 1) caused by biochar applications. According to the Failed Safe N-test, the significant N₂O reduction is robust against a possible positive publication bias because a huge number of non-significant observations would need to be furthermore included in the meta-analysis (>650,000) to turn the significant result into a non-significant result (Table S4). However, biochar induced reductions of N₂O emissions were of transient nature with a tendency to be negligible within one year (Fig. S1). In spite of a non-significant overall effect on NO₃⁻ leaching, biochar significantly and consistently reduced NO₃⁻ leaching by 26 to 32% in studies with an experimental time of >30 days (Fig. 1). In parallel, available NO₃⁻ in soils decreased over time with significant reductions for experiments conducted for >120 days (Fig. 1).

3.2. Biochars

Results suggest a dependency of biochar feedstock selection and conversion technology on N dynamics (Fig. 2). Biochars produced of wood and lignocellulosic biomass by gasification, slow pyrolysis, and their combination with steam at each heating temperature, reduced soil N₂O emissions ($P < 0.05$). On the other hand, N₂O emissions remained unaffected after application of i) biochars made of manure and biosolids ($P = 0.095$) and ii) biochar produced by fast pyrolysis (particle residence time a few seconds; Bruun et al. 2012) and iii) hydrochars produced via hydrothermal carbonization (Libra et al.

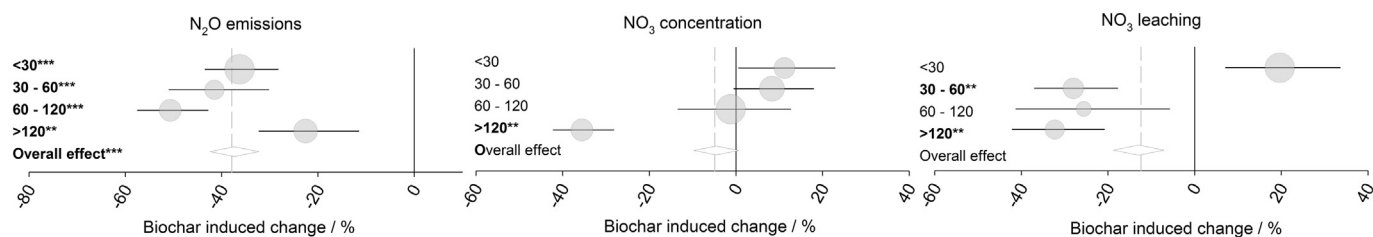


Fig. 1. Impact of study length (days) on soil N_2O emissions, NO_3^- concentration in soil after study, and NO_3^- -leaching during study. Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (***) for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$).

2011). Concentrations of NO_3^- significantly decreased in soils amended with biochars produced at heating temperatures $<500^\circ\text{C}$ and with those produced by fast pyrolysis. Biochars produced from lignocellulosic biomass and biochars produced at temperatures of $>500^\circ\text{C}$ reduced NO_3^- leaching.

Particle size of biochar particles did not affect N_2O emissions, NO_3^- concentration, and NO_3^- leaching (NO_3^- leaching_{fine biochar}: $P = 0.069$; Fig. 3). N_2O emissions were reduced after biochar application over a broad range of biochar pH, N and C contents, except for biochars consisting of $<460 \text{ g C kg}^{-1}$; a relatively small number of biochar types (5.9 to 12.8 g N kg^{-1} and C/N mass ratio of 100 to 200) reduced the NO_3^- concentration in soil. Exclusively wood biochars characterized by pH values ranging between 7.8 and 8.9 increased NO_3^- concentration in soil, while acidic to neutral ($\text{pH} < 7.8$) and strongly alkaline ($\text{pH} > 9.6$) biochars reduced leaching of NO_3^- . Leaching of NO_3^- was further reduced by biochars consisting of $<780 \text{ g C kg}^{-1}$, N ranging between 3.3 and 5.9 g kg^{-1} , and C/N mass ratio of 100 to 200.

3.3. Soils

Soil N_2O emissions were reduced regardless of soil texture (i.e. clay, silt, sand) as reflected by small data variability ($-34 \pm 8\%$; strongest reduction in soil with $>70\%$ sand: -47%) (Table S4). The concentration of NO_3^- in soil varied strongly ($-6 \pm 26\%$) among grouped textures (Table S4) and only coarse textured soils (i.e. sand) showed a reduced concentration and leaching of NO_3^- (Fig. 4).

Biochar reduced N_2O emissions at each soil pH and C/N ratio (Fig. 5). However, in soils with SOC concentrations $>24 \text{ g kg}^{-1}$ and total N concentrations $>3 \text{ g kg}^{-1}$, the N_2O emission reduction was smallest and not significant. Furthermore, biochar applications reduced soil NO_3^- concentrations in slightly acidic to neutral soils ($\text{pH} 5.5$ to 7.0), in soils

that contained low SOC concentrations ($<10 \text{ g kg}^{-1}$), and in low total N soils ($<1 \text{ g kg}^{-1}$). Interestingly, soils that were less affected by biochar in terms of soil NO_3^- concentrations actually showed reduced soil NO_3^- leaching ($\text{pH} < 5.5$; $10\text{--}24 \text{ g C kg}^{-1}$, $0.7\text{--}1.7 \text{ g N kg}^{-1}$, C/N mass ratios of >9.3 and >12.4).

3.4. Soil types and soil management

Man-made soils (i.e. Anthrosols represented in this study exclusively by paddy soils), organic soils (i.e. Histosols), sandy soils (i.e. Arenosol), and soils typical for steppe and sub-humid temperate climate (i.e. Luvisol) showed reduced N_2O emissions after biochar applications (Figs. 7 and 8). Biochar applications reduced NO_3^- concentration only in Luvisols (i.e., soils of sub-humid temperate climate). Soil NO_3^- leaching was exclusively reduced in Cambisols (i.e. soils of limited age), and semi-arid soils (e.g. Calcisol, Solonetz) (Figs. 6 and 7).

Low biochar application rates of $<10 \text{ Mg ha}^{-1}$ neither affected N_2O emissions nor NO_3^- leaching, but increased NO_3^- concentration in soils (Fig. 8). Larger biochar application rates (e.g., $>10\text{--}20 \text{ Mg ha}^{-1}$) reduced N_2O emissions, and tended to reduce NO_3^- leaching and concentration. N_2O emissions, NO_3^- concentrations in soils, and NO_3^- leaching remained unaffected for soils managed by application of biochar in combination with organic fertilizers. Compared to unfertilized soils, the N_2O emission mitigation potential of biochars was larger for fertilized soils (reduction of -46% for mineral fertilizer [e.g. NH_4NO_3 , $(\text{NH}_4)_2\text{SO}_4$, KNO_3], -34% for urea, -32% for mixtures of organic and mineral fertilizers, and -27% for unfertilized soils; Fig. 8). The impact of biochar and fertilizers on NO_3^- concentration in soils varied, with reduced NO_3^- concentration for biochar experiments fertilized with mineral N and increased NO_3^- concentration in soils after use of urea in combination with biochar. Additions of organic fertilizer and

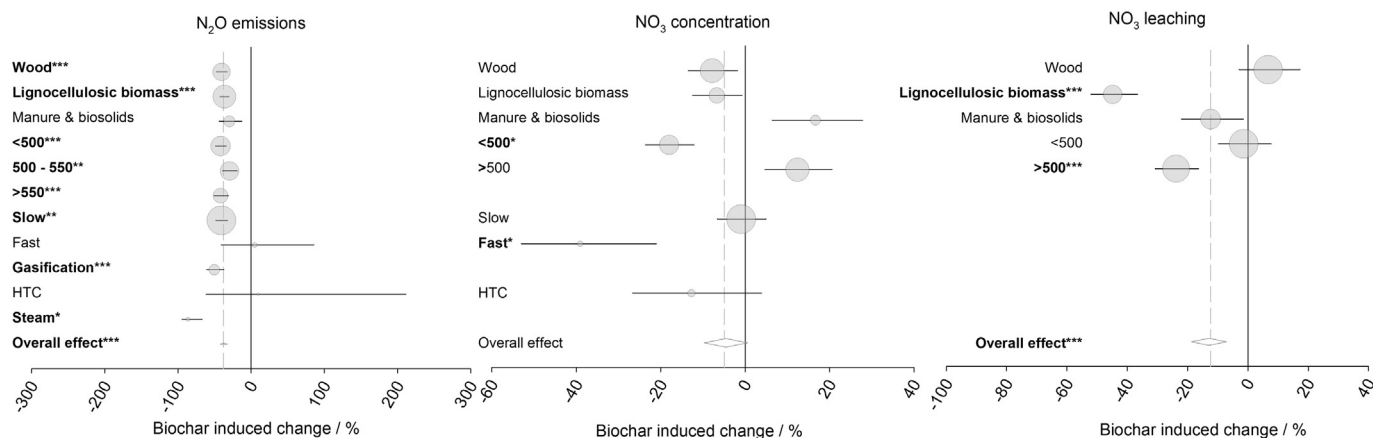


Fig. 2. Impact of feedstock (wood, lignocellulosic [i.e. all other non-woody] biomass, and manure & biosolids), process temperatures ($<500^\circ\text{C}$, 500 to 550°C , $>550^\circ\text{C}$), and production technologies (slow pyrolysis, fast pyrolysis, gasification, hydrothermal carbonization [HTC], and steam) on soil N_2O emissions, NO_3^- concentration in soil after study, and NO_3^- -leaching during study. Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (***) for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$).

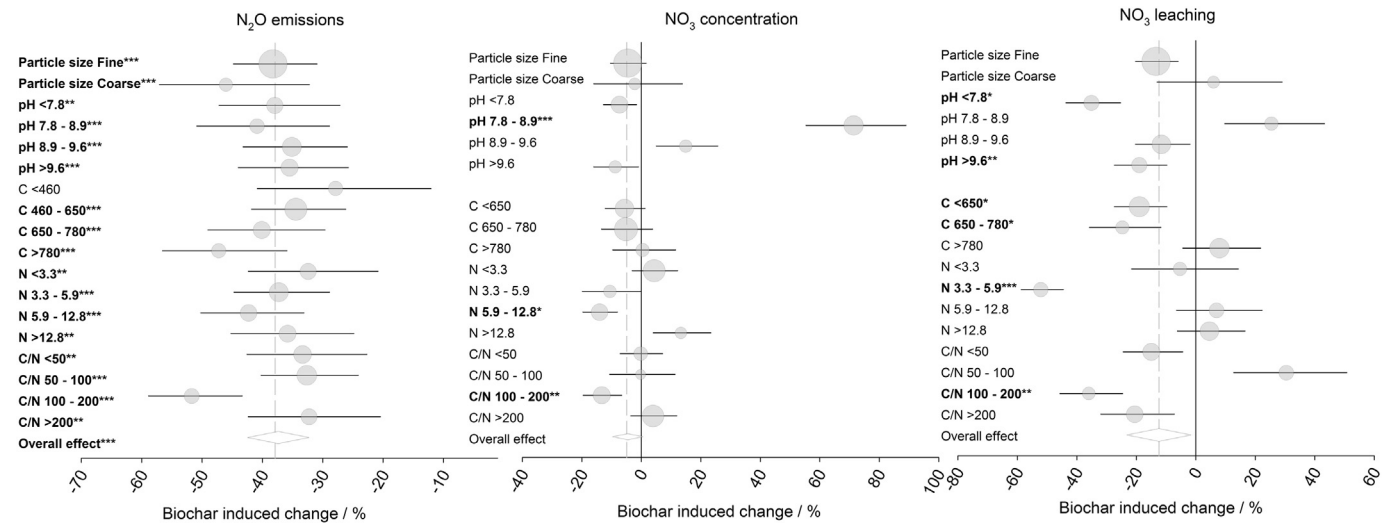


Fig. 3. Impact of various biochar properties (particle size, pH, carbon [C; g kg⁻¹], nitrogen [N; g kg⁻¹], and C/N mass ratio) on soil N₂O emissions, NO₃⁻ concentration in soil after study, and NO₃⁻-leaching during study. Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (*** for P < 0.001; ** for P < 0.01, and * for P < 0.05).

unfertilized biochar experiments did not affect NO₃⁻ concentration measured after experiments. Leaching of NO₃⁻ was reduced by biochar in unfertilized soil and when the fertilizer application rate was below 150 kg N ha⁻¹ (Fig. 8). For each N application rate, biochar induced a reduction of N₂O emissions, but leaching of NO₃⁻ progressively increased in response to increased N application rates (i.e. -26% for <150 kg N ha⁻¹, -7% for 150–300 kg N ha⁻¹, 46% for >300 kg N ha⁻¹). Hence, over all N fertilizer application rates, the NO₃⁻ leaching reduction was not significant (i.e. +3% with mineral fertilizer, -7% with organic fertilizer, and -35% with organo-mineral fertilizer). In parallel to reduced NO₃⁻ leaching in soils fertilized with <150 kg N ha⁻¹, concentration of NO₃⁻ was also reduced. Larger N application rates showed increased concentration of NO₃⁻ after application of 150 to 300 kg N ha⁻¹, but did not affect NO₃⁻ concentration after application of >300 kg N ha⁻¹.

Except for perennial crops (e.g. fruit trees), N₂O emissions at least tended to be reduced in all agronomic experiments (i.e. arable crops with -45% and horticultural cultures with -32%) that cultivated cereals (-31%, P < 0.05), maize (-31%, P = 0.17), rice (-40%, P < 0.05), vegetables (-30%, P = 0.07), and other crops (-35%, P = 0.18) (Fig. 9). Compared to control soils, agricultural soils enriched with

biochar were further depleted in NO₃⁻ while NO₃⁻ leaching was reduced. Biochar applications to grassland increased NO₃⁻ concentration in soils, but neither N₂O emissions nor NO₃⁻ leaching were affected.

4. Discussion

Overall, soil N₂O emissions and NO₃⁻ leaching were reduced after biochar applications, while soil NO₃⁻ concentration remained overall unaffected. These findings are in line with meta-analysis results published by Liu et al. (2018) and Nguyen et al. (2017), except that their findings (based on 796 observations) indicated a significant reduction of 12% in soil NO₃⁻. In the current study, reasons for reduced NO₃⁻ leaching and an at least unaffected concentration of NO₃⁻ in soil were presumably the result of soil processes affected by biochar and the ability of biochar to reversibly take up and release nitrate (Kammann et al. 2015, Haider et al. 2016, 2017; Hagemann et al. 2017a). Soil properties and processes can be modified by biochar in several ways. Biochar increases soil pH due to its “liming effect” (Clough et al., 2013; Hüppi et al., 2015; Nguyen et al., 2017), which induces a shift of the NH₄⁺/NH_{3(g)} equilibrium promoting release of NH₃ at elevated pH values, biochar-induced NH₃ volatilization, particularly from acidic soils, may

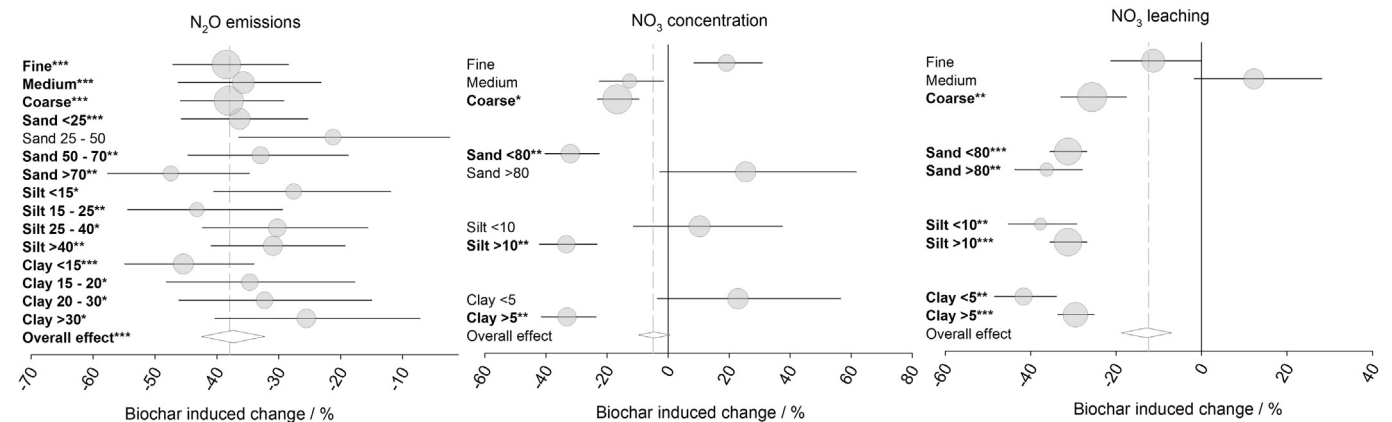


Fig. 4. Biochar effects on soil N₂O emissions, soil NO₃⁻ concentration post-study, and NO₃⁻-leaching during study affected by soil texture classes (fine or clay, medium or silt, coarse or sand textured) and proportion (%) of sand, silt, and clay. Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (*** for P < 0.001; ** for P < 0.01, and * for P < 0.05).

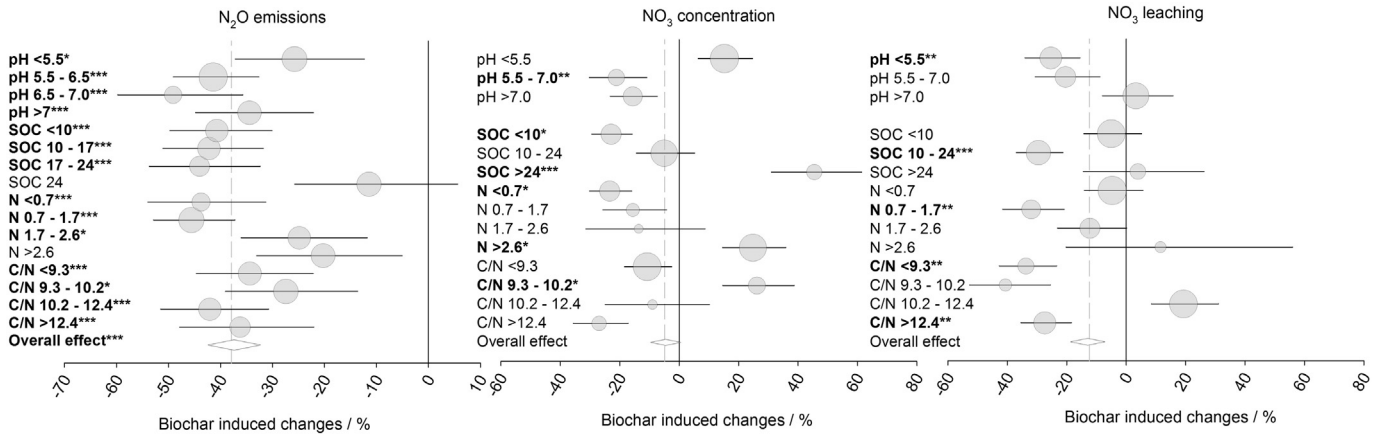


Fig. 5. Biochar effects on N_2O emissions, soil NO_3^- concentration post-study, and NO_3^- leaching during study affected by chemical soil properties (pH, soil organic carbon [SOC; $mg\ kg^{-1}$] and nitrogen [N; $mg\ kg^{-1}$], C/N mass ratio). Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (**** for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$).

have reduced soil NO_3^- concentration and leaching (Liu et al., 2018). Other potential mechanisms explaining NO_3^- concentration and leaching from soil are i) a presumed sorption of NO_3^- on biochar (Nguyen et al., 2017; Sumaraj and Padhye, 2017; Yao et al., 2012), ii) biochar entrapment of NO_2^-/NO_3^- (Kammann et al. 2015; Haider et al., 2016, 2017; Hagemann et al., 2017a, b; Pignatello et al., 2006), and iii) stabilization of NO_3^- in biochar pores and organo-mineral coatings on biochars that hampers release, but promotes nitrate capture (Kammann et al., 2015; Hagemann et al., 2017a, b; Haider et al., 2016; Joseph et al. 2017). Thus, biochar can alter processes controlling NO_3^- formation substantially, but its interaction with pathways of NO_3^- stabilization and retention remain elusive.

The same mechanisms may further explain reduced N_2O emissions, with multiple processes needing to be considered to explain biochar-induced alteration of N_2O emissions. It has been shown that biochar addition can promote the expression of the N_2O reductase genes of denitrifiers (*NosZ*), promoting a complete reduction of NO_3^- to N_2 instead of N_2O (Wang et al., 2013; Harter et al., 2014), thereby shaping microbial communities (Harter et al., 2016a, b; Krause et al., 2018). Theoretically, the effect can be related to a slight pH increase around biochar particles; it is well known that a higher soil pH promotes a more complete denitrification to N_2 . On the other hand, a reduction of the nitrate concentration around biochar particles can reduce the concentration of “ NO_3^- substrate” for denitrifiers which would be in line with the observation of an increase in the *NosZ* gene expression. Furthermore, one may assume that biochar adds a fraction of readily degradable organic C sources (Lan et al., 2017; Wang et al., 2016), which induces a reduction of N_2O emissions (Barnard et al., 2005; Cayuela et al., 2013; Lan et al.,

2017) by microbial growth and N immobilization. These processes impart a complex study matrix upon systems that receive biochar, which may explain the lack of process-based knowledge in the literature.

4.1. Impact of time

Our meta-analysis indicated a transient nature of biochar on N_2O emissions reduction, which needs more research regarding the mechanisms, and which needs to be taken into account as a dynamic factor when assessing biochars' long-term greenhouse gas mitigation potential (Woolf et al., 2010). Factors that affect biochars' long-term ability to reduce soil N_2O emissions includes biochar aging within soil, leading to biochar surface property changes via oxidation and formation of oxygen-containing functional groups (Mia et al., 2017), sorption of natural organic matter leading to clogged biochar pores (Kasozi et al., 2010; Pignatello et al., 2006; Sumaraj and Padhye, 2017; Hagemann et al., 2017b), and formation of organo-mineral complexes coating biochar surfaces (Joseph et al., 2017; Sumaraj and Padhye, 2017). Alteration of surface functional groups will affect electrostatic interactions, reducing the capacity of biochar to sorb NO_3^- (Güereña et al., 2012; Mia et al., 2017; Nguyen et al., 2017) while organo-mineral complexes tend to increase NO_3^- retention by mechanisms still under debate (Hagemann et al., 2017b; Joseph et al., 2017; Nguyen et al., 2017; Sumaraj and Padhye, 2017). Hence, aging of biochar and capturing NO_3^- may explain progressive reduction of NO_3^- concentration in and leaching from soil, which can suppress N_2O emissions from denitrification pathways (Bouwman et al., 2002; Cayuela et al., 2013; Pelster et al., 2011).

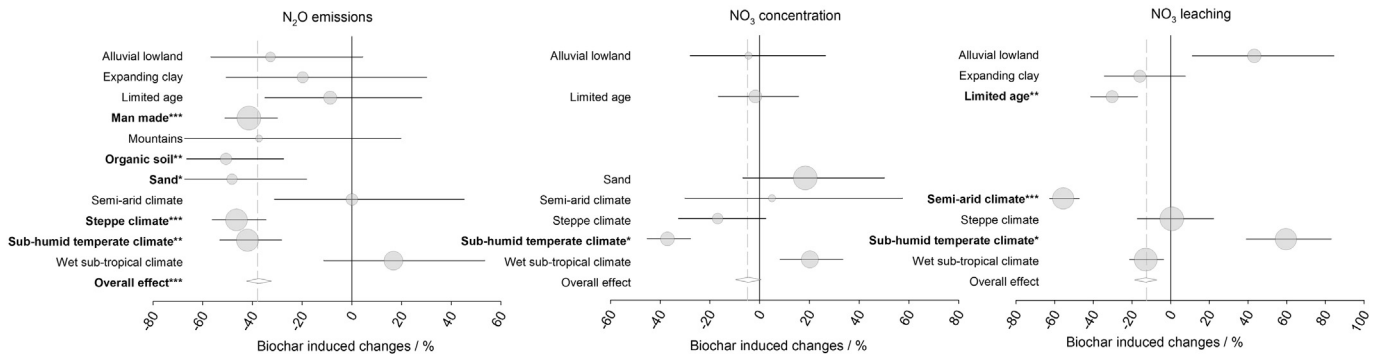


Fig. 6. Biochar effects on N_2O emissions, soil NO_3^- concentration post-study, and NO_3^- leaching during study affected by soil classes (Driessen et al., 2001). Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (**** for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$).

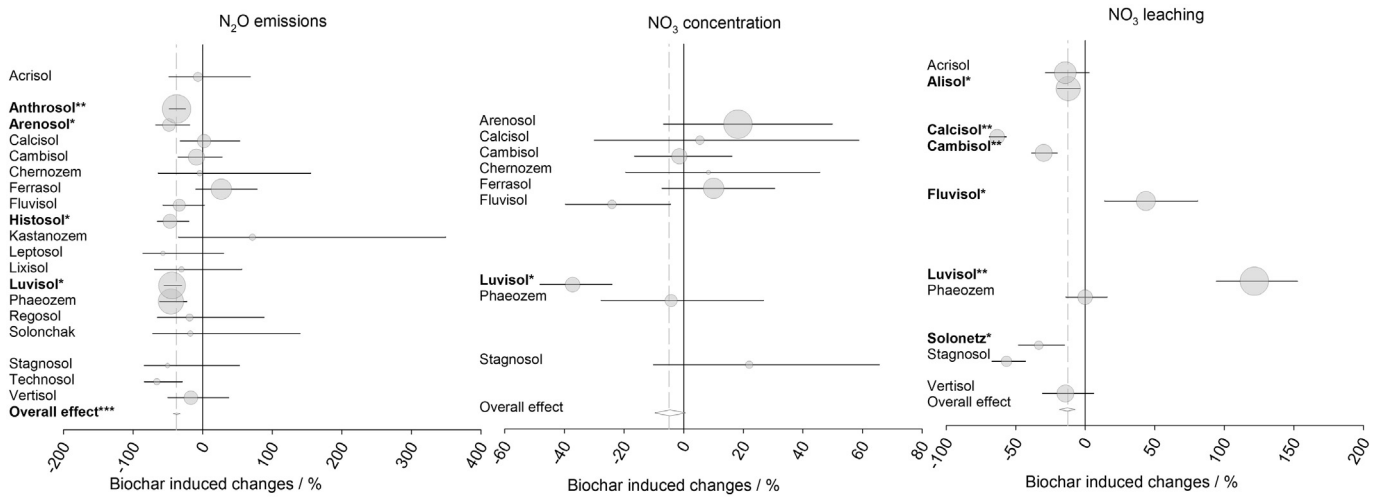


Fig. 7. Biochar effects on N_2O emissions, soil NO_3^- concentration post-study, and NO_3^- leaching during study affected by soil types (IUSS Working Group WRB, 2014; see USDA counterparts below figure). Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (*** for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$). WRB = USDA: Acrisol = Ultisol; Alisol = all 12 USDA soil orders; Anthrosol = Inceptisol; Arenosol = Inceptisol/Entisol; Calcisol = Aridisol; Cambisol = Inceptisol; Chernozem = Mollisol; Ferrasol = Oxisol; Fluvisol = Entisol; Histosol = Histisol; Kasatanozem = Mollisol; Leptosol = Alfisol; Luvisol = Alfisol; Phaeozem = Mollisol; Regosol = Entisol; Solonchak = Aridisol; Solonetz = Aridisol; Stagnosol = Alfisol/Ultisol/Entisol/Mollisol; Technosol = Anthrosol (man-disturbed); Vertisol = Vertisol.

4.2. Impact of biochar production and properties

Results suggested that N_2O emissions remained unaffected after application of N-rich biochars (e.g., manure and biosolids feedstocks) characterized by C/N ratios similar to soil organic matter ($\text{C/N}: 17 \pm 1$), while application of N-poor biochars made from wood and lignocellulosic biomass ($\text{C/N}: 279 \pm 13$ and 112 ± 5 , respectively) reduced N_2O emissions, similar to the findings of Liu et al. (2018). Our results further indicate that NO_3^- concentration remained overall unaffected, but Liu et al. (2018), who utilized a larger number of observations, revealed a significant decrease in soil NO_3^- concentration (12%). Our study revealed that NO_3^- concentrations were reduced at N fertilizing rates ($<150 \text{ kg N ha}^{-1}$) typically applied to biochar experiments as also reviewed by O'Connor et al. (2018b), but increased when N fertilization was higher ($150\text{--}300 \text{ kg N ha}^{-1}$). Similar to previous research (e.g., Clough et al., 2013; Kanthle et al., 2016; Demiraji et al., 2018; Liu et al., 2018), soil NO_3^- leaching was overall reduced by biochar

application. Factors that may explain these findings are the availability of easily biodegradable organic C and ash present in biochar, both affecting soil N transformation or nitrate capture by biochar. Biochar produced at temperatures $>500 \text{ }^\circ\text{C}$ are almost free of labile organic C while those produced at temperatures $<500 \text{ }^\circ\text{C}$ can contain labile organic material (Keiluweit et al., 2010; Wang et al., 2016; Zimmerman et al., 2011); biochar-borne labile organic C sources may induce microbial immobilization of N or capture NO_3^- by organic coating found on aged biochars (Borchard et al., 2014c; Clough et al., 2013; Hagemann et al., 2017a, b). Applied biochar particles that contain ash are slightly alkaline (i.e. close to 7.8) and provide a spatially limited, but optimal environment for substantially stimulated nitrifier activity (Antoniou et al., 1990; Barnard et al., 2005; Nguyen et al., 2017), which may explain elevated NO_3^- concentrations in soils amended with biochars having pH between 7.8 and 8.9. Another potential pathway for a stimulated nitrifier activity is the sorption of inhibitory phenolic compounds that can reduce or block nitrifier activity e.g. in acidic conifer forest soils (DeLuca et al. 2006).

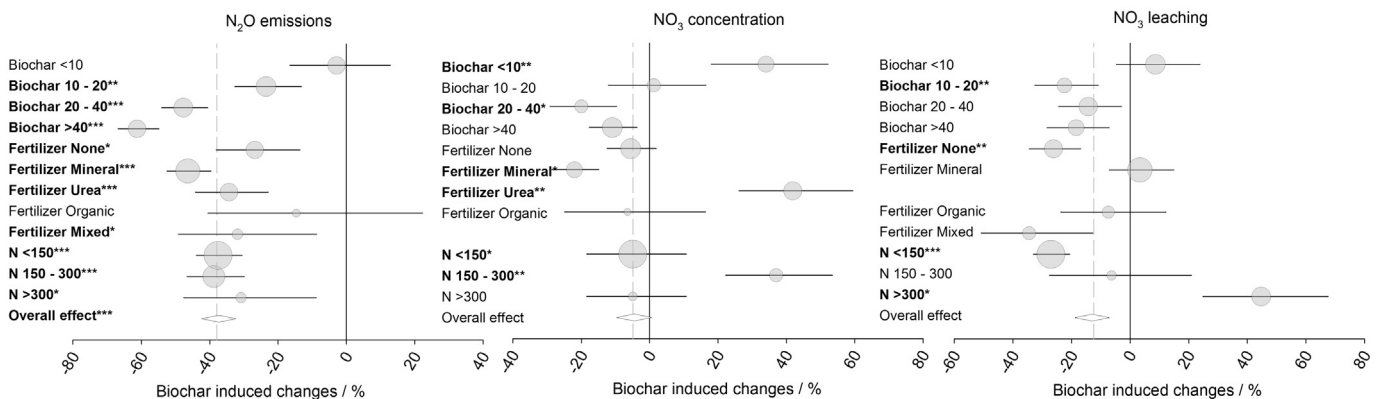


Fig. 8. Biochar effects on N_2O emissions, soil NO_3^- concentration post-study, and NO_3^- leaching during study affected by soil amendments (i.e. biochar application rate [Mg ha^{-1}], fertilizer type and N application rate [kg ha^{-1}]). Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (*** for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$).

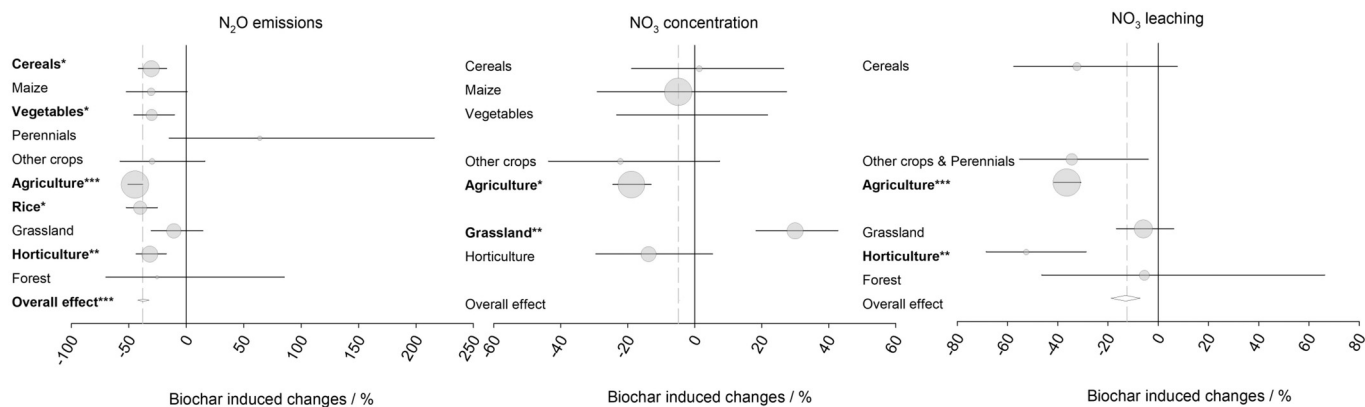


Fig. 9. Biochar effects on N_2O emissions, soil NO_3^- concentration post-study, and NO_3^- leaching during study affected by cultivated crops (cereals, maize, rice, vegetables, perennials, and other) and land use type (agriculture, grassland, horticulture, and forest). Data are shown as estimated mean effects and their lower and upper confidence intervals (95%). Circle size indicates number of observations (see also Supplementary information). Solid vertical line indicates mean of control treatments and dashed vertical line indicates mean of overall effect. Probability levels are indicated by asterisks (***) for $P < 0.001$; ** for $P < 0.01$, and * for $P < 0.05$.

4.3. Impact of soil type and soil properties

Biochar applications increased NO_3^- concentration in very coarse soils (>80% sand) and soils poor in organic C and N. Although the exact mechanisms are unknown, this could be a result of biochar application that increased total soil organic C and a stimulated stabilization of inherent soil C (Borchard et al., 2014a; Hernández-Soriano et al., 2015; Kasozi et al., 2010; Pignatello et al., 2006), a subsequent accelerated N transformation, and sorption and/or retention of NO_3^- (Nelissen et al., 2012; Clough et al., 2013; Liu et al., 2018). Adding biochar to coarse textured soils typically affects water retention and flow (Ajayi and Horn, 2016; Clough et al., 2013; Petersen et al., 2016), which is assumed to reduce NO_3^- leaching as stated by Liu et al. (2018) and this meta-analysis. When applied to acidic soils and soils rich in soil organic matter, biochar may have induced decomposition of native soil organic matter (Ding et al., 2017; Wang et al., 2016), stimulating soil organic matter mineralization, nitrification, and formation of NO_3^- (e.g. Nelissen et al., 2012). In comparison to findings by Liu et al. (2018), our meta-analysis results showed that N_2O emissions remained unaffected in soils rich in organic matter (>24 g kg^{-1} , here exclusively grassland soils as compared to other agricultural soils, see below) at elevated NO_3^- concentration in soil stimulating an incomplete reduction of NO_3^- (Barnard et al., 2005); moreover, the complete reduction of N_2O to N_2 during denitrification was suppressed by low pH-values of these soils. Thus, pH, organic matter and texture are controlling factors explaining biochar-induced formation and retention of NO_3^- in soil and corresponding N_2O emissions.

Soils that are well aerated, coarse textured, and typically poor in soil organic matter are Arenosols, while Histosols are rich in organic matter yet may also be anoxic (Driessen et al., 2001). Our results revealed that biochar applications to Arenosols had no effect on NO_3^- concentration and leaching (-14%), but biochar reduced N_2O emissions by -48%. Arenosols are promoting almost complete nitrification of NH_4^+ to NO_3^- (Chapin et al., 2011), which biochar evidently enhances in Arenosols (Meusel et al., 2018; Nguyen et al., 2017). Soils naturally known to be a source of N_2O emissions are Histosols and paddy soils (i.e. man-made soils or Anthrosols) that emit N_2O , especially during transitions of dry-to-flooded or flooded-to-dry soil conditions due to seasonally varying water regimes (Dalal et al., 2003; Kögel-Knabner et al., 2010; Liu et al., 2010; Peng et al., 2011; Schwärzel et al., 2002). Our results revealed that biochar applications to Histosols reduced N_2O emissions by -47%. This N_2O emissions reduction may be improved by biochar through i) alteration in soil moisture regime during non-flooded periods (Ajayi and Horn, 2016; Clough et al., 2013; Petersen et al., 2016), ii) a reduction of redox potentials to levels that promote formation of

NH_4^+ , or iii) complete denitrification to N_2 (Harter et al., 2014; Barnard et al., 2005; Cayuela et al., 2013; Sumaraj and Padhye, 2017).

4.4. Impact of soil conditioner and land use type

Agricultural land use requires replacement of nutrients by fertilization and can mitigate climate change by sequestering C in soils (Werner et al., 2018; Wollenberg et al., 2016). Biochar is thought to sequester C in soils and stabilize non-charred soil organic C (Abdelrahman et al., 2018; Borchard et al., 2014a; Wollenberg et al., 2016), which may have an impact on nitrification and subsequently on N_2O emissions (Nelissen et al., 2012). Our meta-analysis results showed that typical biochar application rates of >10 $Mg\ ha^{-1}$ (ranges typically between 5 and 50 $Mg\ ha^{-1}$; Liu et al., 2013; Lorenz and Lal, 2014) reduced N_2O emissions, while NO_3^- concentration and leaching tended to be reduced, similar to results of Liu et al. (2018). Our findings confirm that biochar-induced alterations of mineral N transformations are dose-related as also shown for crop yields and soil organic C dynamics (Crane-Droesch et al., 2013; Ding et al., 2017). Moreover, biochar applications significantly suppressed N_2O emissions typically induced by N fertilization (Barnard et al., 2005) indicating biochar could be a valuable mitigation tool to lower emission factors of N fertilizers. Assuming maintained or even increased yields after biochar application (Crane-Droesch et al., 2013), an important factor to consider is that reduced N_2O emissions also reduce greenhouse gas emission equivalents per kg produced agricultural crop.

Based on our meta-analysis results, it appears that a fertilizer dose-related mechanism progressively reduced soil NO_3^- leaching and increased soil NO_3^- concentration; this response, however, dropped to a non-significant response at N fertilizer application rates of >300 $kg\ ha^{-1}$. This effect likely may be due to the limited capacity of biochars to immobilize and entrap NO_3^- (Borchard et al., 2014b; Clough et al., 2013; Hagemann et al., 2017b), explaining the reduced or even maintained NO_3^- leaching for experiments that received <300 $kg\ N\ ha^{-1}$ compared to those receiving less N fertilization, and increased NO_3^- concentration after application of >150 $kg\ N\ ha^{-1}$. However, biochar could not prevent leaching of NO_3^- after application rates of >300 $kg\ N\ ha^{-1}$. Thus, applying >300 $kg\ N\ ha^{-1}$ to biochar amended soils increases the risks associated with N loss by NO_3^- leaching rather than by N_2O emissions; mechanisms controlling this finding remain unclear.

Patterns of reduced N_2O emissions, NO_3^- concentration and leaching suggest that biochar application reduces N losses from agriculture (i.e. cereals, rice), except for grassland, perennial crops (e.g. grapevine and fruit trees), and forest. Compared to other agricultural soils (pH: $6.3 \pm$

0.1; C: $17 \pm 1 \text{ g kg}^{-1}$), grassland soils (pH: 5.3 ± 0.1 ; C: $34 \pm 1 \text{ g kg}^{-1}$) contained larger stocks of soil organic C, which is in line with current knowledge (Guo and Gifford, 2002; Scharlemann et al., 2014). These differences explain unaffected N_2O emissions and NO_3^- leaching, but increased concentration of NO_3^- in these soils as biochar can induce decomposition of soil organic C (Ding et al., 2017; Wang et al. 2016). The mineralization of soil organic matter can accelerate subsequent nitrification and formation of NO_3^- (Nelissen et al., 2012). Simultaneously, biochar elevates soil pH, which alters the $\text{NH}_4^+/\text{NH}_3(\text{g})$ equilibrium and typically stimulates a more complete reduction of N_2O to N_2 (Clough et al., 2013; Hüppi et al., 2015; Obia et al., 2015; Nguyen et al., 2017; Liu et al., 2018), which was assumed to reduce N_2O emissions. Obviously, our knowledge of biochar-induced effects on soil N_2O emissions especially on grassland and perennial crops is incomplete.

5. Conclusion

This meta-analysis revealed that biochar stimulates an overall N_2O emissions reduction of 38% with greater reductions immediately after application. The time dependent impact of biochar application on soil N_2O emissions is a crucial factor requiring consideration in order to develop and test resilient and sustainable biochar-based greenhouse gas mitigation strategies. In terms of land use, biochar can reduce rice paddy soil N_2O emissions (i.e. Anthrosols) by almost 40%; this may significantly mitigate climate change, as ~140 Mha are used as paddy fields globally (Kögel-Knabner et al., 2010), and since methane emissions from paddy soils have been found to be reduced as well (Jeffery et al., 2016). Adding biochar to sandy or coarse textured soils (e.g. Arenosols) reduced both N_2O emissions and NO_3^- leaching, which reduces soil N losses and presumably improves both N use efficiency and mitigates climate change. Considering land use (e.g., paddy soils, grasslands, annual or perennial cropping systems, etc.) in conjunction with soil properties (e.g. texture, soil organic matter, pH) may provide reliable information suitable for up-scaling N_2O emission reduction estimations and potentials, and ultimately the best practical scenarios for environmental biochar use. Our results support the notion of a dose-response relationship of biochar application on N_2O emission reduction and also NO_3^- leaching, which hints towards the interesting possibility of using biochar as a carrier matrix for “carbon-fertilizers” as successfully explored by Qian et al. (2014). Using biochar in this way would greatly reduce the required dose of N per hectare, which would improve N use efficiency and reduce the economic barriers for biochar use in agronomy. However, the eco-physiological mechanisms controlling N uptake by plants in soil-biochar-plant systems require further analyses to ensure sustainable N-management.

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