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A Meta-Analysis Study on the Use of Biochar to Simultaneously Mitigate Emissions of Reactive Nitrogen Gases (N_2O and NO) from Soils

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Abstract: Nitrous oxide (N_2O) and nitric oxide (NO) are detrimental reactive gaseous oxides of nitrogen. Excessive application of nitrogen fertilizers in cropping systems has significantly increased the emissions of these gases, causing adverse environmental consequences. Previous studies have demonstrated that biochar amendment can regulate soil-N dynamics and mitigate N losses, but they lacked simultaneous assessments of soil N_2O and NO emissions. Thus, the factors influencing the emissions of nitrogen oxides are still unclear. Therefore, this study examined the impact of biochar application on simultaneous N_2O and NO emissions based on 18 peer-reviewed papers (119 paired observations). A machine learning model (boosted regression tree model) was adopted to assess the potential influencing factors, such as soil properties, biochar characteristics, and field management conditions. The addition of biochar reduced N_2O and NO emissions by 16.2% and 14.7%, respectively. Biochar with a high total carbon content and pH, from woody or herbaceous feedstock, pyrolyzed at a high temperature, applied at a moderate rate and to soil with a high-silt content, a moderate pH, and coarse texture, could simultaneously reduce soil N_2O and NO emissions. Biochar amendment, thus, has the potential to lower the environmental impact of crop production. Furthermore, the influence of soil properties, biochar characteristics, and field management should be considered in the future to enhance the efficacy of biochar.

Keywords: N_2O emissions; NO emissions; biochar; influencing factors

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1. Introduction

Nitrogen (N) fertilizer application can improve crop yield, economic benefits, and ensure food security [1,2]. In the pursuit of high yields, excessive N fertilizer application is prevalent in agricultural practices, resulting in a N surplus in the crop-soil system and causing reactive N (Nr) release into the environment [3,4]. Nitrous oxide (N_2O) and nitric oxide (NO) are detrimental Nr gases, and their emissions have increased. N_2O is an important long-lived greenhouse gas, and its centennial scale global warming potential is 265 times higher compared to CO_2 [5–9]. NO is a key substance in tropospheric chemistry, participating in complex photochemical reactions and contributing to acid rain and secondary inorganic aerosol formation [9–11]. Soil nitrification and denitrification are the primary production pathways for NO and N_2O emissions. Thus, the simultaneous monitoring of these trace N-gases is important to understand the soil-N cycle. To simultaneously mitigate NO and N_2O release, relevant influencing factors must be understood; the "hole-in-the-pipe" model suggests that factors which control the N transformation rate and number of oxidized/reduced molecules produced are key to regulating NO and N_2O emissions [12].

Biochar is a carbon-rich material, with a high-ash content, alkalinity, and stability [13,14]. Biochar amendment in arable soils is a promising technique and has shown great potential to improve carbon sequestration and pollutant reduction [15,16]. Additionally, introducing external C would change the composition of microbial communities and the abundance of functional genes, and subsequently affect the soil-N cycle, including nitrification and denitrification [17,18]. Biochar amendment might enhance soil adsorption capacity due to its fine surface porosity and functional groups and then substantially limit the availability of NH_4^+-N which is a substrate for N losses [19–21]. Previous studies have widely reported significant N_2O reduction after biochar amendment, and the relevant influencing factors were determined [18]. The "hole-in-the-pipe" model revealed a trade-off effect for these trace N-gas emissions, suggesting that high NO emissions may relate to lower N_2O emissions. Therefore, after biochar amendment, it is essential to evaluate NO and N_2O emissions simultaneously. Factors which affect biochar efficacy including biochar characteristics, as well as soil properties and field management practices need to be further investigated.

Therefore, this study collected published research on simultaneous NO and N_2O observations after biochar application; analyzed simultaneous reduction potential; and further interpreted the effects of soil properties, biochar characteristics, and field management on the reduction efficiency, using a meta-analysis combined with machine learning models. The aim of the study was to promote biochar application and provide recommendations for mitigating N oxide emissions, which would reduce environmental impacts.

2. Materials and Methods

2.1. Database Compilation

In this study, peer-reviewed scientific papers published from 1970 to 2022 were obtained from the Web of Science, Scopus, and the China National Knowledge Infrastructure (CNKI) database; the keywords used for the search were 'biochar', 'black carbon', 'N_2O emissions', and 'NO emissions'. Duplicate data were removed by screening experimental details including location, year, and author. Articles were then selected using the following criteria:

(I) The study must have a paired experiment design, with a treatment group (biochar amendment) and control group (without biochar), both of which had similar field management (including fertilization, irrigation, and tillage regimes).
(II) Soil N_2O and NO emissions were monitored simultaneously.
(III) The mean and standard deviation values of the cumulative emissions of the two N oxides, as well as the number of replicates, were documented.

A total of 18 peer-reviewed articles with 119 paired observations met the selection criteria, and the detailed selection procedure referred to PRISMA statement (Figure S1). Data were collected from these articles and extracted by using a digitized tool in Origin 2019 (Northampton, Massachusetts, USA). The soil properties, biochar characteristics, and field management conditions were also collated. Soil properties included: pH, organic carbon (SOC, g kg^{-1}), total nitrogen (g kg^{-1}), clay (%), silt (%), sand (%), and texture. Biochar characteristics included: pH, application rate (BCAR, t ha^{-1}), total carbon content (TC, %), total nitrogen content (TN, g kg^{-1}), source feedstock, and pyrolysis temperature (PT, °C). Field management conditions included: experiment type, fertilizer type, and nitrogen application rate (NAR, kg N ha^{-1}). The details of each were categorized as follows:

(A) Soil properties:
- (i) Soil pH: pH $\leq$ 6; 6 < pH $\leq$ 8; and pH > 8.
- (ii) Soil organic carbon g kg^{-1}: SOC $\leq$ 10; 10 < SOC $\leq$ 20; and SOC > 20.
- (iii) Total nitrogen g kg^{-1}: TN $\leq$ 1; 1 < TN $\leq$ 2; and TN > 2.
- (iv) Clay %: clay $\leq$ 30 and clay > 30.
- (v) Silt %: silt $\leq$ 30 and silt > 30.
- (vi) Sand %: sand $\leq$ 30 and sand > 30.

(vii) Soil texture: fine (sandy clay, silty clay, and clay); medium (sandy clay loam, clay loam, and silty clay loam); and coarse (sandy, loamy sand, sandy loam, loam, silt loam, and silt) [21].

(B) Biochar properties:

(i) Biochar pH: pH $\leq$ 7; 7 < pH $\leq$ 9; and pH > 9.
(ii) Biochar application rate t ha^{-1}: BCAR $\leq$ 10; 10 < BCAR $\leq$ 20; and BCAR > 20.
(iii) Total carbon %: TC $\leq$ 45; 45 < TC $\leq$ 65; and TC > 65.
(iv) Total nitrogen g kg^{-1}: TN $\leq$ 6; 6 < TN $\leq$ 12; and TN > 12.
(v) Pyrolysis temperature °C: PT $\leq$ 400; 400 < PT $\leq$ 500; and PT > 500.
(vi) Feedstock types: herbaceous waste; wood; and manure.

(C) Field management conditions:

(i) Experimental types: field; greenhouse; pot; and incubation.
(ii) Fertilizer types: urea; ammonium; compound; and organic.
(iii) N Application rate kg N ha^{-1}: NAR $\leq$ 200; 200 < NAR $\leq$ 400; and NAR > 400.

2.2. Meta-Analysis

In this study, the natural logarithmic response ratio ln(RR) was used to calculate the magnitude of the effect of biochar amendment in each study by using the following calculation formula [22]:

$$\ln(RR) = \ln\left(\frac{X_T}{X_C}\right) \tag{1}$$

where X_T is the cumulative N_2O or NO loss in biochar amendment treatment, and X_C is the cumulative N_2O or NO loss in the control (without biochar amendment). The magnitude of the effect of each study was weighted by the inverse pooled variance [22,23]:

$$V_i = \frac{{S_T}^2}{N_T {X_T}^2} + \frac{{S_C}^2}{N_C {X_C}^2} \tag{2}$$

where N_T and N_C are the sample sizes of the treatment and control groups, respectively, and S_T and S_C are the standard deviations of the treatment and control groups, respectively.

The mean effect of N_2O and NO emissions and confidence interval (CI) of each subgroup was calculated using a hierarchical mixed-effect model, which included the random effects among different studies and study sampling variations [24,25]. Restricted maximum likelihood was used to estimate each parameter in the meta-analytical models. Heterogeneity of effect size (Qm) was also calculated to assess the influence among different groups. Egger's regression test was used to examine publication bias of the meta-analysis [26] (Table S1).

The subgroup analysis mainly considered the effects of soil and biochar properties, and field management conditions on the efficacy of biochar amendment to mitigate N oxide emissions. A meta-analysis was conducted using R 4.1.3 (University of Auckland, New Zealand) and 'Metafor' package. The results (mean effect size and its 95% CI) were then converted to percentage change using the formula: %change = $(e^{\ln(RR)} - 1) \times 100$ [13]. Negative or positive values indicated that the biochar amendment decreased or increased N oxide emissions, respectively, after biochar amendment. The effect of biochar amendment on N oxides emissions was considered significant if the 95% CI did not overlap with the zero line. The subgroups significantly differed if the 95% CI did not overlap with each other. The results were presented in forest plots which are drawn using 'ggplot2' package in R 4.1.3 (University of Auckland, New Zealand).

2.3. Analysis of Influencing Factors Affecting Biochar Amendment Efficacy

Traditional linear regression cannot process complex non-linear relationships between response variables and explanatory variables. Therefore, a boosted regression tree (BRT) analysis and machine learning models were adopted to rank the variable's influence on

N oxide emissions after biochar amendment [13,27]. The BRT model fits thousands of simple models to describe the relationship between explanatory variables and response variables (effect size), and then summarizes the results of multiple single tree models using the boosting method. This improves the performance and predictive ability of the model, and avoids model overfitting [28,29]. Additionally, the BRT model can manage missing values in the dataset [30]. The model in R4.1.3 (University of Auckland, New Zealand) uses the 'gbm', 'dismo', and 'ggBRT' packages [31,32]. The model was established using the "Gaussian" error structure, and a 10-fold cross-validation was adopted to assess predictive error of individual models [33]. The parameters were obtained by systematically adjusting the learning rate (0.1, 0.005, and 0.001) and bag fraction (0.5, 0.6, and 0.75). A final learning rate (0.001) and bag fraction (0.75) were chosen [34]. The BRT model analysis shows that the factors with significant contributions can be analyzed and screened, based on the number of explanatory variables [32]. Subsequently, the correlation between response and explanatory variables was determined by fitting a linear mixed-effect model (for continuous variables only).

3. Results

3.1. Effects of Biochar Application on Soil-Nitrogen Oxide Emissions

Biochar application to agricultural soil simultaneously reduced the emissions of N_2O (RR: −16.2%, CI: −29.6 ~ −0.2%) and NO (RR: −14.7%, CI: −24.8 ~ −3.4%) (Figure 1A). There was no significant difference in the molar ratio of NO and N_2O between the amendment and control groups (Figure 1B).

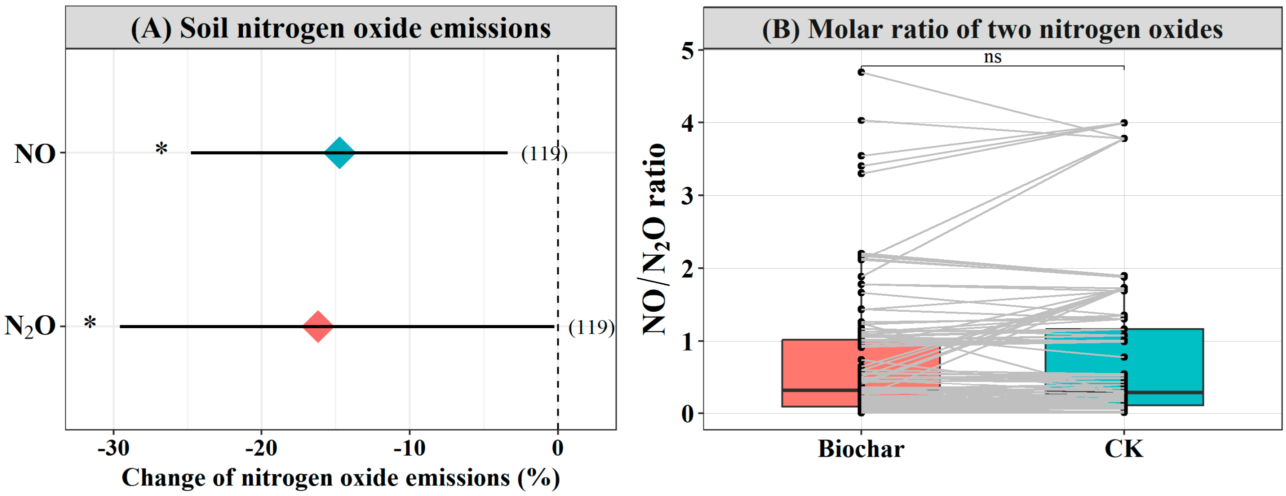

Figure 1. Effects of biochar amendment on soil−nitrogen oxide emissions. (**A**) Changes in emissions of the two N oxides after biochar amendment. The numbers on the right indicate the number of observations and the asterisks (*) on the left indicate significant effects of biochar amendment. (**B**) Mole ratio of NO and N_2O emissions between the biochar amendment and control. The difference between biochar and CK was evaluated using student's *t*-test, and ns indicated no significant difference.

3.2. Effects of Soil Properties on Nitrogen Oxides Mitigation Potential of Biochar Amendment

Biochar applied to acidic and alkaline soils had no significant effect on N_2O and NO emissions. However, in neutral soils ($6 < pH \leq 8$), the emissions were reduced significantly (Figure 2). N_2O (RR: −36.7%, CI: −50.1~−19.7%) and NO (RR: −42.7%, CI: −54.3~−28.2%).

N_2O emissions were further significantly reduced by adding biochar to soils with moderate organic carbon content ($10 < SOC \leq 20$), high-silt content, and a lower sand and clay content (RR: −29.1%, CI: −44.9 ~ −8.8%). Results showed that soils with a higher silt content or coarse texture significantly reduce N_2O and NO emissions simultaneously.

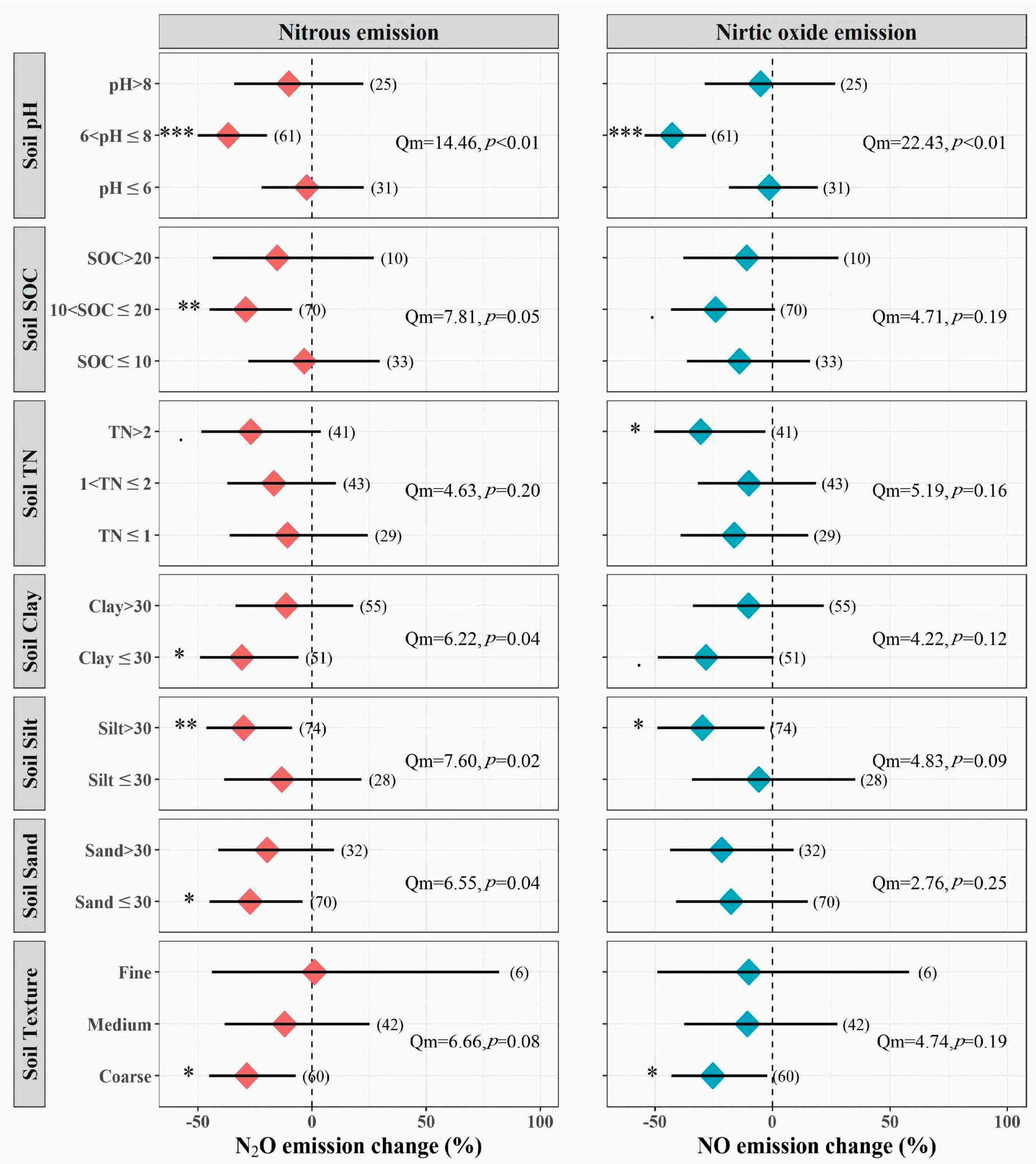

Figure 2. Effects of soil properties on soil−nitrogen oxide emissions after biochar amendment. The asterisks indicate significant effects of biochar amendment, with *, **, and *** indicating significance levels at 0.05, 0.01, and <0.01, respectively.

3.3. *Effects of Biochar Characteristics on Nitrogen Oxide Mitigation Potential of Biochar Amendment*

A high biochar application rate (>20 t ha^{-1}) reduced NO emissions by 25.3%, while a moderate rate (10 < BC AR ≤ 20 t ha^{-1}) resulted in further reduction for both N_2O (RR: −29 %, CI: −45.6~−7.4%) and NO (RR: −22%, CI: −38.3~−1.2%) (Figure 3). Additionally, biochar with a high pH and total carbon content, pyrolyzed at high temperature or produced using herbaceous waste or wood, also significantly reduced both N_2O and NO emissions. NO emissions also decreased after the application of biochar pyrolyzed at moderate temperature (400 < PT ≤ 500 °C), or with moderate pH, and using biochar with a relatively high (TN > 12) or low (TN ≤ 6) total nitrogen content (TN).

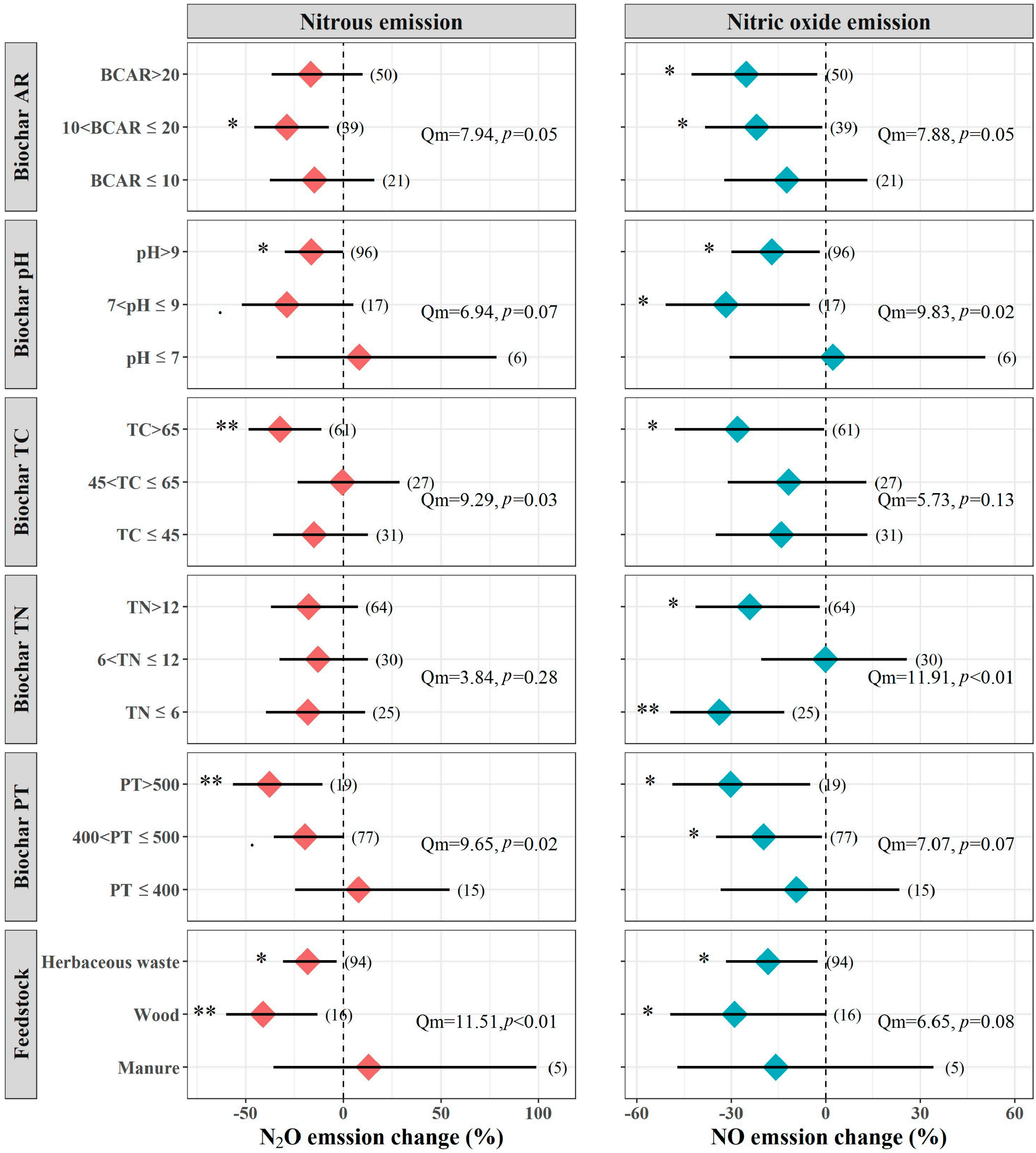

Figure 3. Effects of biochar characteristics on soil-nitrogen oxide emissions after biochar amendment. The asterisks indicate significant effects of biochar amendment, with *, ** indicating significance levels at 0.05, 0.01, respectively.

3.4. Effects of Field Management Conditions on the Nitrogen Oxide Mitigation Potential of Biochar Amendment

Experiments were conducted under different field conditions; however, no differences were observed. Applying urea (RR: −20%, CI: −34.3~−2.3%) or compound fertilizer (RR: −18.3%, CI: −33~−0.5%) with biochar amendment can significantly reduce N_2O emissions. However, using ammonium fertilizer had the opposite effect (Figure 4). Fertilizer types had little impact on NO emissions, but a relatively low N application rate ($N \leq 200$ kg N ha^{-1}) reduced 23.2% of NO emission compared to that from the control sample (no biochar amendment)

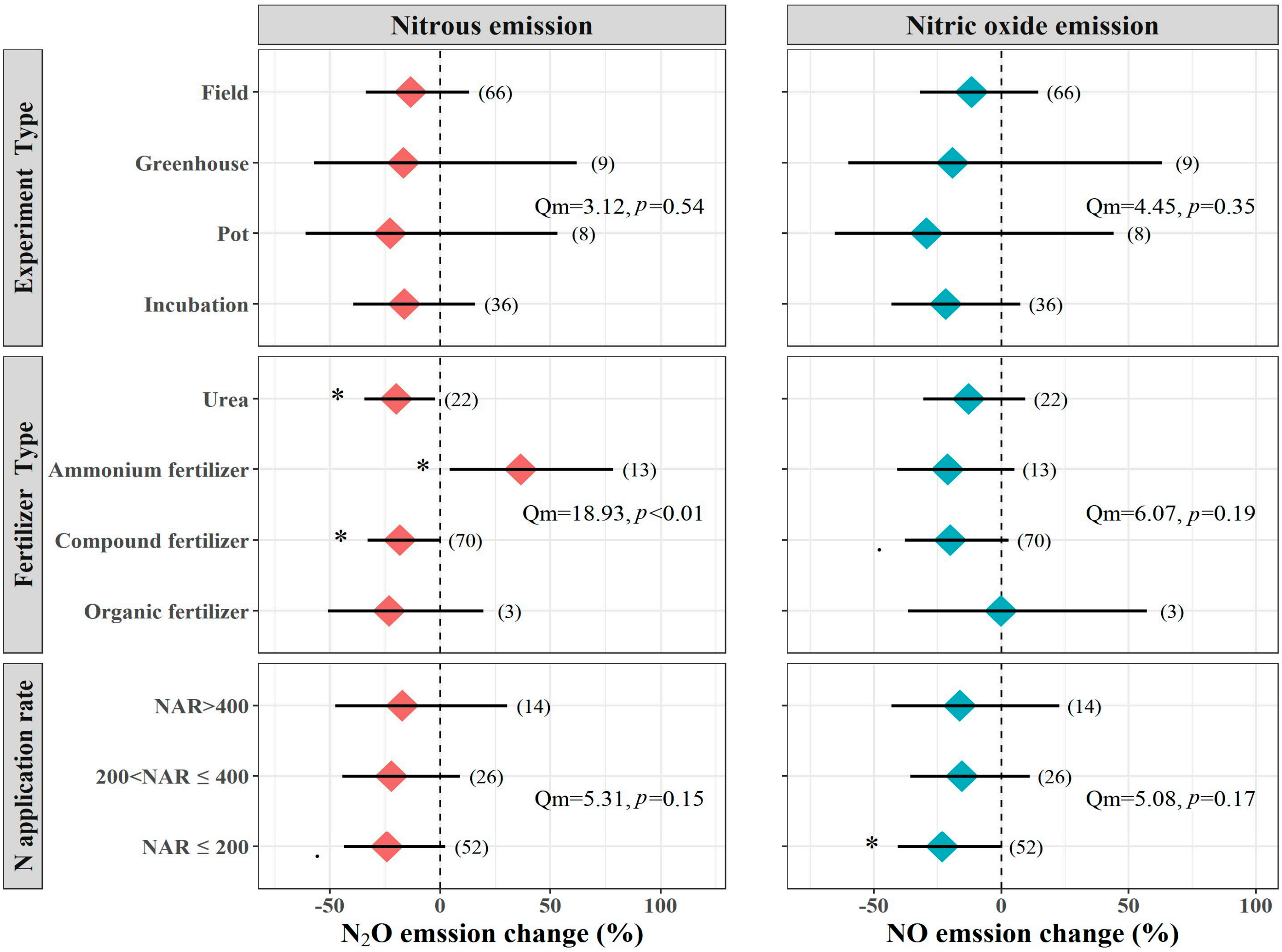

Figure 4. Effect of field management conditions on soil-nitrogen oxide emissions after biochar amendment. The asterisks indicate significant effects of biochar amendment, with * indicating significance levels at 0.05, respectively.

3.5. Influencing Factors Affecting Soil-Nitrogen Oxide Emissions after Biochar Amendment

Based on the results of the BRT model, total carbon content of biochar, soil-silt content, fertilizer type, and nitrogen application rate had a significant effect on the N_2O emission after biochar addition (Figure 5A). Soil-silt content and biochar application quantity exerted a significant impact on NO emissions. Combined with the mixed-effect model, the increase in the total carbon and pyrolysis temperature of biochar enhanced its N_2O and NO emission mitigation potential. The change in silt content showed a similar trend; however, the biochar pH change showed the opposite correlation (Figure 5 and Figure S1).

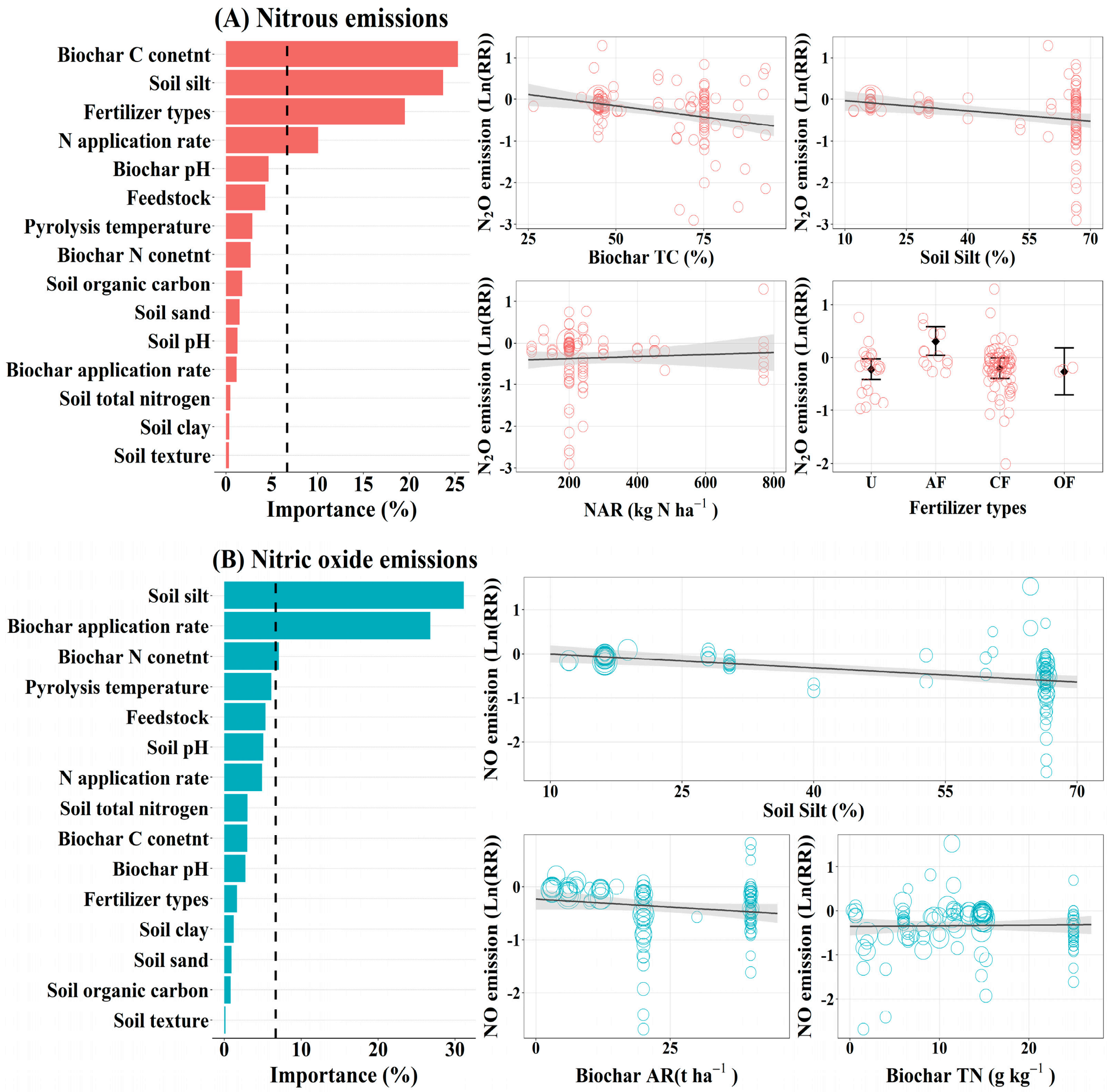

Figure 5. Analysis of influencing factors affecting soil−nitrogen oxide emissions after biochar amendment. (**A**) N_2O emissions and (**B**) NO emissions. The importance of each influencing factor (Figures on the left) was obtained using the boosted regression trees model (BRT), the importance crossing the black dash line denotes the factors significantly impacting the effect size. A mixed-effect model (Figures on the right) was adopted to describe the relationship of effect size with each influencing factor; effect sizes were weighted (bubble sizes).

4. Discussion

4.1. Effects of Soil Properties on N_2O and NO Emissions after Biochar Amendment

The results of the meta-analysis showed that biochar amendment significantly reduced N_2O and NO emissions from fertilized agricultural soils; however, the potential for reduction varied depending on the characteristics of the soil, biochar, and field management conditions. The BRT model indicated that soil texture contributes significantly to changes in nitrogen oxide emissions after biochar application to the soil. In contrast to fine textured soils, biochar added to coarsely textured soil resulted in higher N_2O and NO reduction potential, which may be attributed to the soil's enhanced nutrient-retention capacity [35]. Nitrogen oxide emissions correlated negatively with soil-silt content. High-silt content soils are often associated with poor aeration and thus a higher denitrification process related to nitrogen oxide emissions. However, biochar application can improve soil aeration due to its porous structure and large specific surface area, thereby inhibiting the denitrification derived N_2O and NO emissions [36,37]. Soil with a low-clay content can enhance the cation exchange capacity and ammonium adsorption by soil, thereby reducing nitrification substrates and N losses [38,39].

Soil pH was also a key factor affecting the soil-nitrogen dynamics [40,41]. The results showed that biochar application to soil with moderate pH ($6 < pH \leq 8$) had a significant impact on N_2O and NO reduction, in line with the study by Cayuela et al. (2014) [36]. Ammonia oxidation was found to be the first and critical rate-limiting step for nitrification. Ammonia-oxidizing bacteria (AOB) carrying the *amo*A gene play an important role in N losses from soil. Biochar addition to relatively neutral soils reduced AOB significantly, thereby suppressing nitrification [42]. However, due to the "liming effect" of biochar addition, the increased soil pH in an aerobic micro-environment can stimulate the activity of nitrogen oxide reductases (*nor*B and *nos*Z) and result in complete denitrification to nitrogen gas (N_2) [36,43,44]. Xiao et al. (2019) [42] found that biochar amendment increased the *nos*Z gene abundance in moderately alkaline soil. The effect of soil moisture on NO and N_2O emissions cannot be ignored; when the soil water filled pore space (WFPS) is less than 60%, the NO:N_2O molar ratio is typically greater than one, indicating that the N_2O production pathway is dominated by nitrification. However, higher soil WFPS would lead to a greater denitrification contribution to N_2O emissions and an NO:N_2O ratio lower than one [45–47]. In this study, the NO:N_2O molar ratio did not change much after biochar amendment, but previous studies have confirmed that biochar addition increased the field water capacity by 20.4%, with some variations based on the soil texture, biochar porosity, and surface area [48,49].

4.2. Effects of Biochar Characteristics on N_2O and NO Emissions

In comparison to manure biochar, the application of biochar derived from woody or herbaceous feedstocks can decrease N_2O and NO emissions significantly because they generally have a higher C/N ratio. This leads to N immobilization and subsequent reduction in the substrates in the soil for nitrification-denitrification processes [40,50–52]. Additionally, a larger surface area, more developed pore structure, and a higher degree of aromatization were observed in woody biochar. Aromatic structures with conjugated π-electron system can act as an "electron shuttle" and facilitate the redox reactions of N-cycle functional microorganisms [18,53,54], thereby suppressing N emissions.

Biochar that has been pyrolyzed at high temperatures has higher redox activity and, consequently, redox-active moieties on the biochar surface [55]. Chen et al. [53] found the electron accepting quinones, dominant in high-temperature produced biochar, decrease N_2O emissions. On the other hand, using such biochars would promote N_2O and NO reduction by increasing enzyme activities, such as nitrous oxide reductase (NOS) and nitric oxide reductase (NOR) [43,56]. This further supports the negative correlation between pyrolysis temperature and N oxide emissions. However, the intensity of C=O bond stretching was high in low-temperature pyrolyzed biochar due to the broken conjugated π bonds, which may impede its ability to transfer electrons [53,54,57]. Additionally, the acid functional groups decreased, and alkaline functional groups increased on the biochar surface, resulting in an increase in pH [58,59]. This also created a favorable environment for the stability of nitrite and limited its chemical decomposition to NO [17]. This further confirmed that the application of high-pH biochar is an effective measure to reduce soil-N oxide emissions.

Biochar application rate was also a vital indicator affecting soil-N oxide emissions. By using the X-ray photoelectron spectroscopy technique, Quin et al. [60] observed a significant adsorption of N_2O by surface functional groups (C=C) of biochar and increasing the amount of biochar adjusted the redox potentials, favoring the reduction in N_2O to dinitrogen (N_2). A relatively high biochar addition rate would also create a N-limiting soil environment, inhibiting the substrate source for net nitrification [47,52].

4.3. Effects of Field Management Conditions on N_2O and NO Emission after Biochar Amendment

N fertilization induced pulse emissions of N_2O and NO, which were generally positively related to the N input rate [61,62]. The reduction efficiency of biochar amendment was low in high-fertilizer input cases, which may be related to insufficient adsorption, or a reduction in the capacity of biochar to handle the accumulating N fertilizer in the soil. This can result in substantial amounts of N undergoing nitrification or denitrification [63].

Urea added to the soil is rapidly hydrolyzed to NH_4^+-N by urease, providing sufficient substrate which stimulates the activity of ammonia-oxidizing archaea (AOA), thereby increasing the nitrification rate [64,65]. However, biochar amendment decreased N_2O emissions by improving the soil C/N ratio and decreasing soil-mineral nitrogen availability [41,66,67]. Biochar combined with organic fertilizer had no significant effect on N emissions; however, due to the small sample size of this subgroup, there is a need for further investigation of this aspect. The addition of ammonium fertilizers to soils increased nitrification substrates and promoted nitrification dominated N_2O emissions [2,66]. Mulvaney et al. [68] concluded that the addition of ammonium nitrogen fertilizers promoted the denitrification process by affecting the water-soluble organic carbon content and pH of the soil. Feng et al. [66] demonstrated that the effect of biochar combined with ammonium fertilizer on N_2O depended on TC/IN in the soil. An increase in TC/IN caused the ammonium utilization path to gradually change from nitrification to immobilization. Biochar can increase soil pH [66]. Thus, the application of ammonium fertilizer under high-pH conditions can easily lead to the accumulation of NO_2^-, which reacts with Fe^{2+} in the soil to generate N_2O [69,70].

Our analysis confirmed that biochar can mitigate N_2O and NO emissions simultaneously under certain conditions. For agricultural applications, it is recommended to use moderate biochar ($10 < \text{BC AR} \leq 20$ t ha^{-1}) and fertilizer ($\leq$200 kg N ha^{-1}) application rates. Biochar pyrolyzed at relatively higher temperature ($\geq$500 °C) have a strong redox capacity and should be considered to reduce the emissions of N_2O and NO. However, there was limited information about the mitigation efficiency of biochar produced under extremely high pyrolysis temperatures (>650 °C), which tend to have a larger surface area and a higher pH, and this should be further studied [56]. Silty soils are usually weakly structured and compacted easily [71], so biochar amendment would improve soil quality and reduce Nr emissions [37]. However, there is still considerable uncertainty about biochar-induced mitigation of N emissions, which deserves further research to advance the use of biochar for nitrogen oxides emission reduction.

5. Conclusions

In this study, the meta-analysis method was combined with the BRT model and used to analyze simultaneous soil N_2O and NO emissions after biochar application. The contributions of soil properties, biochar characteristics, and field management conditions to this process were further evaluated. Overall, biochar addition significantly reduced N_2O (16.2%) and NO (14.7%) emissions simultaneously from soil. The total carbon content of biochar, soil-silt content, fertilizer type, and nitrogen application rate were the main factors affecting N_2O emissions. However, soil-silt content, biochar application rate, and total nitrogen content were the main factors affecting NO emissions. This study comprehensively determined the simultaneous reduction potential of soil N_2O and NO emissions after biochar amendment. It also determined the significant influencing factors impacting the emission process and provides recommendations for future biochar application and management, which will benefit sustainable agricultural development.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su15032384/s1, Table S1: Publication bias of meta-analysis; Figure S1: Literatures retrieval and screening process; Figure S2: Analysis of influence factors of soil nitogen oxides emissions after biochar amendment. (A) N_2O emissions and (B) NO emissions.;

Table S2: List of literature used in the data synthesis. References [72–83] are cited in the supplementary materials.

Author Contributions: Conceptualization, X.Z. and Z.S.; methodology, X.Z.; software, X.Z. and Z.S.; validation, J.L.; formal analysis, X.Z.; investigation, X.Z.; resources, Z.S.; data curation, X.C.; writing—original draft preparation, X.Z.; writing—review and editing, Z.S. and Y.Z.; visualization, S.L.; supervision, Z.S. and Y.Z.; project administration, Z.S.; funding acquisition, Z.S. All authors have read and agreed to the published version of the manuscript.

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