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Biochar Is Comparable to Dicyandiamide in the Mitigation of Nitrous Oxide Emissions from *Camellia oleifera* Abel. Fields

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Abstract: *Research Highlights*: Intensive nitrogen (N) application for agricultural purposes has substantially increased soil nitrous oxide (N_2O) emissions. Agricultural soil has great potential in the reduction of N_2O emissions, and applications of biochar and nitrification inhibitors may be useful for mitigating agricultural soil N_2O emissions. *Background and Objectives: Camellia oleifera* Abel. is an important woody oil plant in China. However, intensive N input in *C. oleifera* silviculture has increased the risk of soil N_2O emissions. As an important greenhouse gas, N_2O is characterized by a global warming potential at a 100-year scale that is 265 times that of carbon dioxide. Thus, mitigation of soil N_2O emissions, especially fertilized soils, will be crucial for reducing climate change. *Materials and Methods*: Here, we conducted an in situ study over 12 months to examine the effects of *C. oleifera* fruit shell-derived biochar and dicyandiamide (DCD) on soil N_2O emissions from a *C. oleifera* field with intensive N application. *Results*: A three-fold increase of cumulative soil N_2O emissions was observed following N application. Cumulative N_2O emissions from the field with N fertilization were reduced by 36% and 44% with biochar and DCD, respectively. While N_2O emissions were slightly deceased by biochar, the decrease was comparable to that by DCD. *Conclusions*: Results indicated that biochar may mitigate soil N_2O emissions substantially and similarly to DCD under specific conditions. This result should be examined by prolonged and multi-site studies before it can be generalized to broader scales.

Keywords: biochar; *Camellia oleifera*; DCD; nitrification inhibitor; nitrous oxide

1. Introduction

Increased atmospheric greenhouse gases (GHGs) as a result of human activities contribute substantially to global warming. Nitrous oxide (N_2O) is an important component of GHGs [1] and is a dominant ozone-depleting substance [2]. Concentrations of atmospheric N_2O increased from 270 ppb in the 18th century to a new high at 329.9 ppb in 2017 [3]. Specifically, the global warming potential at a 100-year scale of N_2O is 265 times that of carbon dioxide [1]. Considering its important role in global warming, reduction of N_2O emissions is crucial for the mitigation of global climate change.

Soil is the largest source of N_2O emissions at 13 Tg N_2O-N $year^{-1}$. Human activities have contributed 7 Tg N_2O-N $year^{-1}$ thus far in the 21st century [4]. Intensive nitrogen (N) applications for agricultural purposes have induced input of 79 Tg synthetic N and 7.4 Tg N of livestock manure per year [5,6]. Therefore, agricultural soil has large potential in the reduction of N_2O emissions and hence for the mitigation of global climate change.

Biochar and nitrification inhibitor applications are useful strategies for N_2O emission mitigation [7–10]. Biochar is produced by slow pyrolysis of organic matter under high temperatures and an anaerobic environment [11]. Biochar application reduced N_2O emissions caused by N fertilization by 33% [7]; this was ascribed to increased soil pH [12] or N immobilization [7]. In addition, 70% of N_2O emissions are emitted from microbial-driven nitrification and denitrification processes [4], which could be effectively inhibited by nitrification inhibitors. Nitrification inhibitors are a class of organic compounds that inhibit the activity of nitrifying nitrifiers, including synthetic nitrification inhibitors such as dicyandiamide (DCD), nitrapyrin, and 3, 4-dimethylpyrazole phosphate, and biological nitrification inhibitors such as methyl 3-(4-hydroxyphenyl) propionate [13] and brachialactone [14]. Nitrification inhibitors reduced N_2O emissions by 44% via inhibition of nitrifying nitrifiers [15]. As a commonly used nitrification inhibitor, DCD deactivates the activity of ammonium monooxygenase enzyme (a copper co-factor enzyme), and hence N_2O emissions [16].

Camellia oleifera Abel. is one of the world's four main woody edible oil crops, with a long cultivation history and wide cultivation area in subtropical China [17] due to the beneficial effects of its oil on human health [18]. *C. oleifera* is mainly cultivated in Typic Hapludult Ultisols (red soil) with lower soil fertility [17,19]. Therefore, intensive N input has been used to increase the yield of *C. oleifera* oil. However, large amounts of N input increase the risk of nitrate N (NO_3^--N) leaching and gaseous N losses, such as N_2O emissions and ammonia volatilization [20,21]. While large amounts of *C. oleifera* fruit shells have been dumped without use, it might be an ideal feedstock for producing biochar for the mitigation of N_2O emissions [22].

Here, we conducted study using biochar derived from *C. oleifera* fruit shells and DCD to examine their effects in the mitigation of N_2O emissions from a *C. oleifera* field with intensive ammonium nitrate (NH_4NO_3) fertilization. We predicted that *C. oleifera* fruit shell-derived biochar or DCD may effectively mitigate soil N_2O emissions.

2. Materials and Methods

2.1. Study Site and Soil Collection

This study was conducted at a *C. oleifera* plantation covering 200 ha in Yongxiu county, Jiangxi province, China (29.16° N, 115.77° E) from 25 February 2017 to 16 March 2018. The *C. oleifera* plantation has been intensively managed more than 10 years, with each individual tree distributed 2 m or 3 m apart. Compound fertilizer with 14% N was applied at the rate of 300 mg $plant^{-1}$. In this region, there is a subtropical monsoon climate with a mean annual precipitation of 1561 mm and a mean annual air temperature of 17.5 °C (the monthly mean temperature ranges from 2.4 °C in January to 33.4 °C in July) (http://www.worldclim.org). Soil was classified as Typic Hapludult (red soil). Soil characteristics were obtained by collecting soil samples from 12 randomly selected sites and pooled together for measurement. The basic characteristics were as follows: bulk density, 1.42 g cm^{-3}; pH, 4.45; total organic carbon (TOC), 11.06 g kg^{-1}; total N (TN), 1.18 g kg^{-1}; dissolved organic carbon (DOC), 0.28 g kg^{-1}; dissolved organic N (DON), 39.78 mg kg^{-1}; ammonium N (NH_4^+-N), 4.52 mg kg^{-1}; NO_3^--N, 1.37 mg kg^{-1}.

2.2. Experimental Design and Field Procedures

This study was conducted using a randomized design with four treatments (including Control, N only, N with Biochar, N with DCD) and four replications (N = 16, four soil amelioration treatment × four replicates). Biochar was produced by pyrolyzing *C. oleifera* fruit shell at 450 °C without oxygen

for 1 h and was applied at the rate of 500 g plant^{-1}(equivalent to 10 t ha^{-1}). Biochar characteristics were: pH, 9.49; TOC, 743.89 g kg^{-1}; TN, 5.14 g kg^{-1}; DOC, 1.57 g kg^{-1}; DON, 14.28 mg kg^{-1}; NH_4^+-N, 2.24 mg kg^{-1}; NO_3^--N, 2.65 mg kg^{-1}. DCD was applied by 2% (DCD/N) [22]. Two years before the study, the studied area was intensively managed but no fertilization was applied. In the study, N was applied by 20 g NH_4NO_3-N plant^{-1} (equivalent to 400 kg NH_4NO_3-N ha^{-1}). Sixteen *C. oleifera* trees with similar size (mean ground diameter: 6.52 cm) were randomly selected and 0.5 m^2 plots were established under the crown of each plant for measurement of N_2O fluxes. Nitrogen, biochar, or DCD were thoroughly mixed and applied in all plots.

Static opaque chamber method was used for measurement of N_2O fluxes. Plastic collars with a groove (inner diameter = 16.7 cm, height = 10 cm, groove = 9 cm) were installed inside each plot. The collar groove was filled with water to seal the open-bottomed chamber (inner diameter = 19.5 cm, height = 80 cm) covered with foam and aluminum for minimizing temperature variation [23]. Gas samples were collected at minutes 0, 5, 10, and 15 min from chamber closing using a syringe, and were stored in aluminum foil gas sample bags before analysis.

Fluxes of N_2O were measured 21 times from 25 February, 2017 to 16 March, 2018 at days 4, 8, 12, 19, 26, 32, 46, 62, 77, 93, 111, 130, 140, 161, 175, 190, 210, 248, 287, 339, and 384. Air temperature, soil temperature, and moisture (10 cm depth) were monitored simultaneously when N_2O fluxes were measured. Meanwhile, soil NH_4^+-N and NO_3^--N (0–20 cm layer) were measured nine times over the study on days 62, 93, 130, 161, 210, 248, 287, 339, and 384.

2.3. Analysis of Soil and Biochar Characteristics

Concentrations of soil and biochar NH_4^+-N and NO_3^--N were extracted by 2 mol L^{-1} KCl solution and measured by a discrete analyzer (Smartchem 200, Rome, Italy). Dissolved organic carbon and DON were extracted by 0.5 mol L^{-1} K_2SO_4 and measured by element analyzer (Multi N/C 3100, Jena Germany). pH was measured by soil (1:2.5, *w/w*) or biochar (1:5, *w/w*) suspensions using pH meter and air-dried samples passed through 0.2-mm sieve (Mettler Toledo, Shanghai, China). Total organic carbon and TN were also analyzed by an element analyzer (Variomax CNS Analyzer, Elementar GmbH, Hanau, Germany) using samples passed through a 0.15-mm sieve.

2.4. Measurement of Soil N_2O Emission Rates and Cumulative Soil N_2O Emissions

Nitrous oxide concentration in each sample was determined using gas chromatograph (Agilent 7890B, Santa Clara, CA, USA). In situ measurements were conducted on sunny days with minimal partial pressure of water vapor. Nitrous oxide fluxes (F, μg m^{-2} h^{-1}) were calculated by [23,24]:

$$F = P \times V \times \frac{\Delta c}{\Delta t} \times \frac{1}{RT} \times M \times \frac{1}{S} \quad (1)$$

where P stands for standard atmospheric pressure (Pa) (which should be adjusted if partial pressure of water vapor of chamber air taken into consideration [25]), V refers to the volume of chamber headspace (m^3), $\Delta c/\Delta t$ means the rate of N_2O (ppb) concentration change with time based on linear regressions [26,27], R stands for universal gas constant (m^3 mol^{-1} K^{-1}), T is the absolute air temperature (K), M means the molecular mass of N_2O (g mol^{-1}), and S indicates the collar area (m^2).

Cumulative soil N_2O emissions (E, μg m^{-2}) were calculated by [28]:

$$E = \sum_{i=1}^{n} \frac{(F_i + F_{i+1})}{2} \times (t_{i+1} - t_i) \times 24 \quad (2)$$

where F indicates soil N_2O emission rates (μg m^{-2} h^{-1}), i means the ith measurement, $(t_{i+1} - t_i)$ refers to the time span (days) between two measurements, and n means the total number of the measurements.

2.5. Statistical Analysis

One-way analysis of variance (ANOVA) was performed to examine dependence of cumulative N_2O emissions on N, biochar and DCD treatments. Repeated-measures ANOVA was used to examine dependence of soil temperature, moisture, NH_4^+-N, NO_3^--N and N_2O emission rates on biochar and DCD treatments. Tukey's honestly significant difference (HSD) tests were used for identifying the significant differences among treatments in ANOVA. Follow-up contrasts were conducted for significant repeated-measures ANOVA results. Pairwise correlation analysis was applied to examine relationship between environment factor, inorganic N and soil N_2O emission rate. All statistical analyses were carried out using JMP 9.0. Software (Gary, NC, USA) at $\alpha = 0.05$.

3. Results

Application of N, biochar, or DCD significantly influenced soil N_2O emission rates ($F = 8.34$, $p = 0.0029$) and cumulative N_2O emissions ($F = 6.68$, $p = 0.0067$) compared to control from the *C. oleifera* field. No significant results were observed in soil temperature, moisture, NH_4^+-N, and NO_3^--N (Figures 1 and 2). Compared with N treatment, N + DCD ($F = 7.94$, $p = 0.0155$) or N + biochar ($F = 5.69$, $p = 0.0344$) treatments showed lower soil N_2O emission rates, but no significant differences were observed between N + DCD and N + biochar treatments ($F = 0.19$, $p = 0.67$; Figure 3). Overall, N, biochar, or DCD treatments significantly impacted soil N_2O emission rates over the 12-month study ($F = 10.11$, $p = 0.0013$; Figure 3).

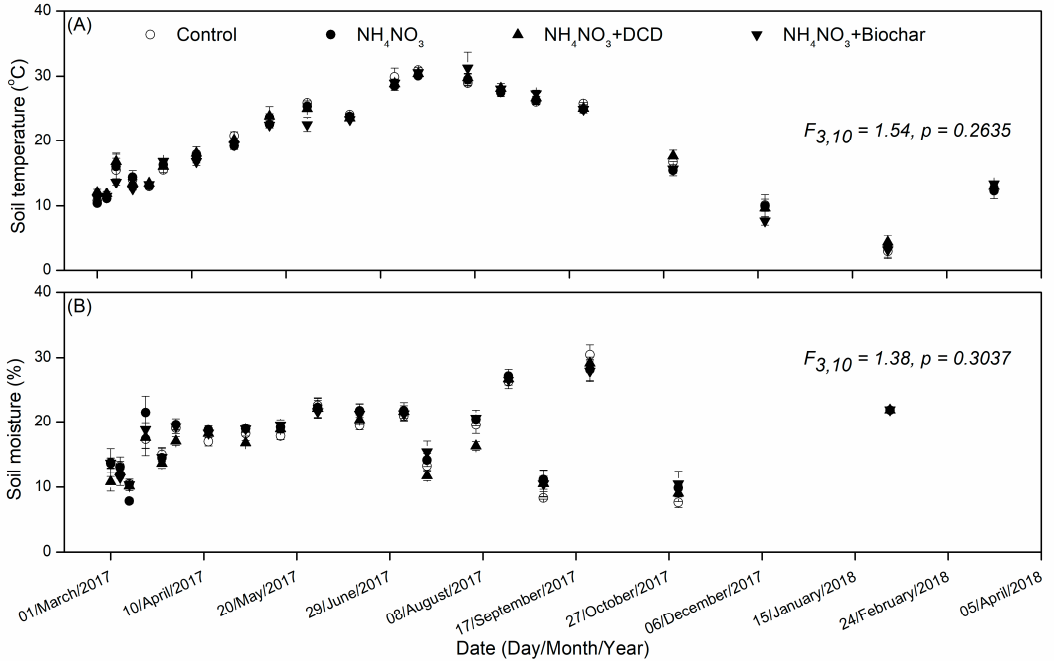

Figure 1. Soil (**A**) temperature and (**B**) moisture (mean ± standard error) over the 12-month study in *Camellia oleifera* Abel. field with the N and mitigation treatments. Repeated-measure one-way analysis of variance results are shown. N: nitrogen; DCD: dicyandiamide; NH_4NO_3: ammonium nitrate.

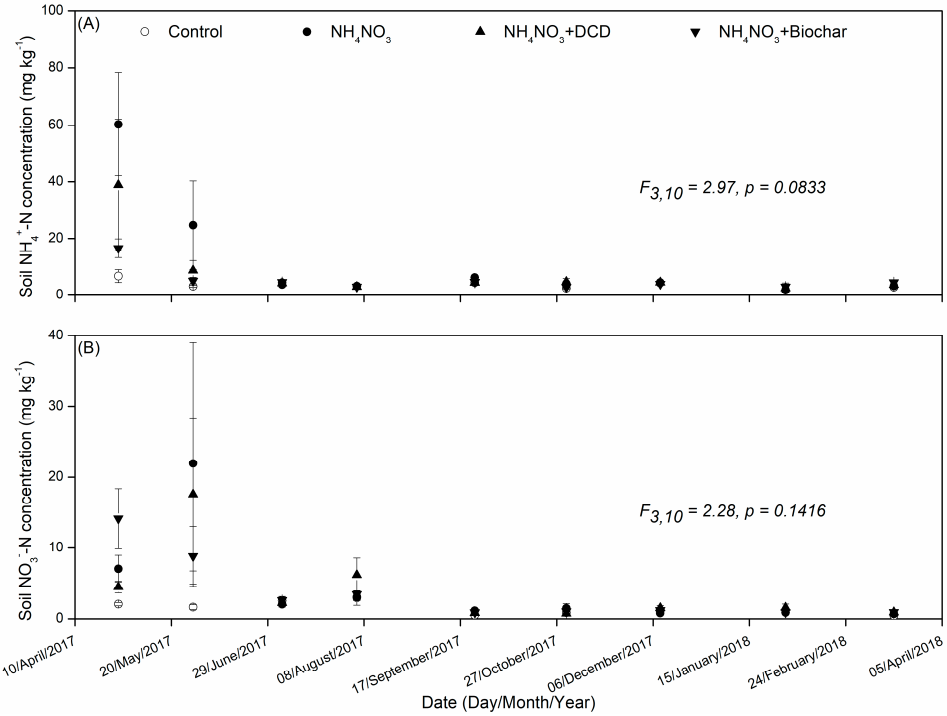

Figure 2. Soil inorganic N dynamics, including (**A**) NH_4^+-N and (**B**) NO_3^--N (mean ± standard error), over the 12-month study in *Camellia oleifera* Abel. field with the N and mitigation treatments. Repeated-measure one-way analysis of variance results are shown. N: nitrogen; NH_4^+-N: ammonium nitrogen; NO_3^--N: nitrate nitrogen; DCD: dicyandiamide; NH_4NO_3: ammonium nitrate.

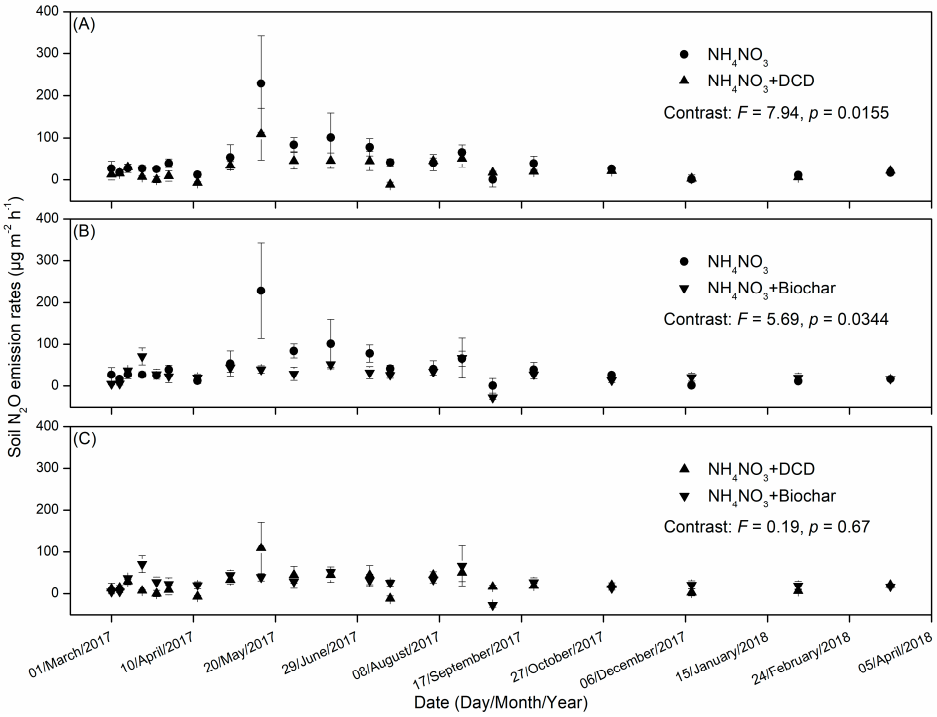

Figure 3. Soil N_2O emissions (mean ± standard error) from soil with N, or N with DCD or biochar in a *Camellia oleifera* Abel. field. (**A**) NH_4NO_3 vs. NH_4NO_3 + DCD; (**B**) NH_4NO_3 vs. NH_4NO_3 + Biochar; (**C**) NH_4NO_3 + DCD vs. NH_4NO_3 + Biochar. Repeated-measure one-way analysis of variance and follow-up contrast results are shown. N: nitrogen; DCD: dicyandiamide; NH_4NO_3: ammonium nitrate; N_2O: nitrous oxide.

Nitrogen treatment increased cumulative soil N_2O emissions (control vs. N, 92.14 ± 47.01 vs. 375.10 ± 60.30 mg m^{-2}, respectively). DCD reduced the increase of cumulative soil N_2O emissions caused by N addition, but no significant differences were observed between N + DCD and N + biochar treatments (Figure 4, N + DCD vs. N + biochar, 211.89 ± 35.88 vs. 238.34 ± 30.65 mg m^{-2}). The soil N_2O emission rate positively correlated with soil temperature, moisture, NH_4^+-N, and NO_3^--N (Table 1).

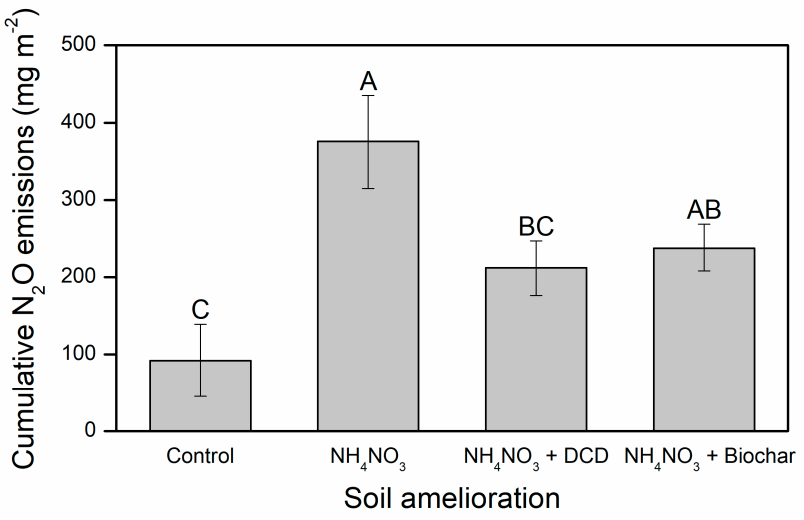

Figure 4. Cumulative soil N_2O emissions (mean ± SE) from the *Camellia oleifera* Abel. field as affected by N fertilization, DCD, or biochar treatments. Bars connected by the same letter are not significantly different in post-hoc tests at $\alpha = 0.05$. N: nitrogen; DCD: dicyandiamide; NH_4NO_3: ammonium nitrate; N_2O: nitrous oxide.

Table 1. Pairwise correlations among soil environmental factors, inorganic nitrogen and soil N_2O emission rate.

Parameters	Soil Temperature	Soil Moisture	NH_4^+-N	NO_3^--N
Soil moisture	0.275 ***			
NH_4^+-N	0.051	−0.050		
NO_3^--N	0.188 *	−0.003	0.414 ***	
N_2O	0.216 ***	0.201 ***	0.285 ***	0.221 **

*, $p < 0.05$; **, $p < 0.01$; ***, $p < 0.001$. NH_4^+-N: ammonium nitrogen; NO_3^--N: nitrate nitrogen; N_2O: nitrous oxide.

4. Discussion

Nitrous oxide emitted from *C. oleifera* plantation was monitored over one year in situ study to investigate effects of biochar or DCD on soil N_2O emissions following application of N fertilization. Soil N_2O emission rates were decreased by biochar or DCD in fertilized soil and the decrease was comparable between two treatments (Figure 3). However, the cumulative soil N_2O emissions caused by NH_4NO_3 fertilization were reduced by DCD application to levels comparable to the control treatment (Figure 4).

4.1. Nitrogen Fertilization Stimulated Soil N_2O Emissions

Nitrogen fertilization stimulated cumulative soil N_2O emissions from *C. oleifera* plantation (Figure 4). N fertilization generally alters activities of N-transforming microorganisms via input of available N substrate [29], stimulating the processes of microbial-driven nitrification and denitrification and subsequent soil N_2O emissions [30,31]. In general, soil N_2O emissions were increased by N input with nonlinear responses [32]. Indeed, the soil N_2O emission rate was positively correlated with NH_4^+-N and NO_3^--N (Table 1). Furthermore, intensive N fertilization, especially NH_4^+-N fertilization, often results in soil acidification [33,34]. Changes in soil pH may regulate soil N_2O emissions via

altering the abundance and composition of N-transforming microorganisms [35–37]. For example, abundances of ammonia-oxidizing bacteria (AOB) were more sensitive to N addition than that of ammonia-oxidizing archaea (AOA) (+ 326% vs. + 27%) [35]. Soil acidification induced by intensive N fertilization results in a high ratio of $N_2O/(N_2O+N_2)$ in the previous study [38]. Therefore, N addition might alter the abundance and composition of AOB and AOA via acidifying soil, hence stimulating N_2O emissions.

In our study, positive correlations between N_2O emission rate and soil temperature or soil moisture were observed (Table 1). A previous study demonstrated that soil temperature and moisture can explain up to 86% variations of N_2O emissions [39]. Soil N_2O emissions varied with soil temperature in specific ranges [28,40], which may relate to different optimum temperatures of N-transforming microorganisms with or without N fertilization and different soil types [37,41]. Compared with soil temperature, soil moisture is the main factor impacting soil N_2O emissions. Consistently, soil N_2O emitted from a wheat–maize plantation showed a positive correlation with a soil water-filled pore space (WFPS) [42]. However, higher soil moisture with lower oxygen content was beneficial to denitrification [30,43] and potentially decrease soil N_2O emissions [44,45]. For example, WFPS at 67–76% was the optimum moisture environment for emitting N_2O [46]. Similarly, N_2O emitted from a rice-rapeseed rotation soil was higher in 60% WFPS than flooding in an incubation experiment [36]. Therefore, moisture effects of soil N_2O emissions may depend on soil type and present non-linear correlations.

4.2. Biochar Reduced Soil N_2O Emission Rates as Affected by N Fertilization

In fertilized soil, N_2O emission rates were significantly decreased and cumulative N_2O emissions were decreased by 36% by biochar (Figures 3B and 4), indicating biochar could be an ideal strategy for N_2O mitigations in *C. oleifera* plantations with N fertilization. Indeed, soil N_2O emissions with N fertilization were decreased 33% by biochar in a meta-analysis study [7]. Biochar-suppressed soil N_2O emissions may be relative, limiting the availability of NO_3^--N to denitrifiers [47,48] or altering the N transformation process rather than limiting the availability of NH_4^+-N or NO_3^--N to N-transforming microorganisms [49]. In addition, biochar could also impose toxic effects on urease activity and subsequent generation of NH_4^+-N by introducing polycyclic aromatic hydrocarbons, heavy metals, and free radicals into soil [50], which may suppress soil N_2O emissions via reducing the N substrate with respect to N-transforming microorganisms.

Biochar addition may suppress soil N_2O emissions by increasing soil pH [12]. The activity of N_2O-reductase was generally higher with higher soil pH [31]. Indeed, the pH of *C. oleifera* fruit shell-derived biochar was higher than that of the acid soil in *C. oleifera* plantations. While an acid soil improvement study showed that liming by dolomite addition could substantially mitigate N_2O emissions via increasing *nosZ* gene abundance [36,51], biochar application could also increase soil pH of the acid *C. oleifera* field soil, which might have also been accompanied by enhanced activities of N_2O-reducing enzymes and hence suppressed N_2O emissions. Moreover, the negative effects of biochar on N_2O emissions could also be induced by its buffer capacity rather than pH, in which biochar acted as "electron shuttle" and replaced NO_3^- as electron sink during denitrification [52]. However, the application of *C. oleifera* fruit shell-derived biochar stimulated N_2O emissions in a previous incubation study [22], which might have been caused by the short-term time scale of incubation study and further indicated the importance of in situ studies. Future studies are still needed for thoroughly understanding of *C. oleifera* fruit shell-derived biochar effects on N_2O emissions and its prolonged effects in mitigation of soil N_2O emissions.

4.3. DCD Reduced Soil N_2O Emissions as Affected by N Fertilization

Cumulative soil N_2O emissions were reduced 44% by DCD application in soil with N fertilization treatment (Figures 3A and 4), which indicated that the application of DCD is an effective strategy for mitigating soil N_2O emissions in *C. oleifera* plantations with intensive N fertilization. DCD has been proved to be effective in reducing average N_2O emission rates following NH_4NO_3 addition in a

previous study [22]. In agreement, DCD reduced soil N_2O emissions following $(NH_4)_2SO_4$ addition by suppressing *amoA* genes and stimulating *nosZ* genes [53]. Nitrification and denitrification are two main pathways producing N_2O [30,31,54]. Application of nitrification inhibitors can suppress soil N_2O emissions [15,16] by inhibiting the activity of ammonium monooxygenase enzyme involved in nitrification process [16]. Thereby, application of DCD generally decreases abundance of *amoA* genes and hence soil N_2O emissions.

4.4. Biochar and DCD Effects on Soil N_2O Emissions

While N fertilization significantly increased soil N_2O emissions compared with control treatment, DCD application decreased soil N_2O emissions to similar levels as control treatment (Figures 3A and 4). Even though biochar addition treatment did not significantly decrease N_2O emissions from soil with N, the slight decrease in cumulative N_2O emissions may potentially mitigate N_2O emissions in prolonged study, which should be examined in future studies. However, DCD application significantly decreased cumulative N_2O emissions and no significant difference was observed between control and DCD treatment (Figure 4), indicating DCD was effective in mitigation of N_2O emissions from *C. oleifera* field relative to biochar. No significant differences were observed between DCD and biochar treatments in their effects on N_2O emission rates (Figures 3C and 4), indicating biochar application could be considered as a potential mitigation strategy of soil N_2O. Similarly, both DCD and biochar reduced the yield-scaled N_2O following N fertilization, while biochar showed stronger effects than DCD in N_2O mitigation in a sweet corn field [55].

5. Conclusions

This study is the first in examining the effects of DCD and biochar derived from *C. oleifera* fruit shells on mitigation of soil N_2O emissions. Application of biochar and DCD showed comparable effects in mitigation of the N_2O emission rate in a *C. oleifera* field with intensive N fertilization practice, with biochar slightly decreasing and DCD significantly decreasing cumulative N_2O emissions. This might have implications for the disposal of dumped byproducts in management of *C. oleifera* and represent an ideal way to enhance both the economic and ecological benefits of the *C. oleifera* industry. If this pattern presents in other plantations, the combined effects of biochar and nitrification inhibitors on soil N_2O emissions should be focused upon in the future. However, the potential effects of biochar derived from *C. oleifera* fruit shell on cumulative N_2O emissions in prolonged studies and other kinds of ecosystems should be examined in future in order to provide guidance for intensive management of *C. oleifera* plantations and disposal of byproducts.

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