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The Effect of Biochar and Straw Return on N_2O Emissions and Crop Yield: A Three-Year Field Experiment

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Abstract: To evaluate the effects of application of biochar and straw return for consecutive years on N_2O emissions and crop yields in North China, a three-year field experiment of applying biochar and straw following a ten-year application was conducted in a wheat–maize rotation system. Four treatments were set up, including F (NPK fertilizer only); FB (NPK fertilizer + 9.0 t·ha^{-1} biochar); FS (NPK fertilizer + straw); and FSB ((NPK fertilizer + 9.0 t·ha^{-1} biochar combined with straw). The results showed that compared with the F treatment, the FB treatment significantly reduced soil N_2O emissions by 20.2%, while the FS and FSB treatments increased it by 23.7% and 41.4%, respectively. The FB treatment reduced soil N_2O emissions by 15.1% in the wheat season and 23.2% in the maize season, respectively. The FS and FSB treatments increased the N_2O emissions by 20.7% and 36.7% in the wheat season, respectively, and by 25.5% and 44.2% in the maize season, respectively. In the wheat season, the soil water content (SWC), NO_3^--N content and pH were the main influencing factors of the soil N_2O emissions. In the maize season, SWC and NO_3^--N content were the main influencing factors. In addition, the FB, FS and FSB treatments increased the crop yield by 4.99%, 8.40% and 10.25% compared with the F treatment, respectively. In conclusion, consecutive application of biochar can significantly reduce N_2O emissions and improve crop yield. Although FS and FSB treatments can also improve the crop yield, they are not beneficial to suppressing N_2O emissions. Therefore, the successive application of biochar is an effective measure to reduce N_2O emissions and maintain crop yield.

Keywords: biochar; straw return; nitrous oxide; nitrate; ammonium

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1. Introduction

The emergence of synthetic nitrogen (N) fertilizer brought some problems while improving crop yield efficiently, and one of those problems is that nitrous oxide (N_2O) emitted from farmland soil through nitrification and denitrification was increased [1]. Compared with carbon dioxide (CO_2), N_2O is at a low level in the atmosphere, while its warming effect within one hundred years is about 300 times that of CO_2, making a significant impact on global climate change [2]. As a major source of N emissions, agricultural soils contributed approximately 33% of global N_2O emissions, and accounted for 62% of N_2O emitted from agricultural activities in China [3]. Therefore, it is of significance to suppress N_2O emissions in the agricultural field to mitigate global climate change.

Many researchers have reported that biochar application and straw return could affect soil N_2O emissions. Biochar is a stable substance that can be derived from the pyrolysis of crop straw and has become a new way of straw utilization. It can improve soil, quality, maintain fertility, and increase crop yield [4,5]. Although biochar has been proved to be beneficial to soil health by many reliable data, there are still some different statements about its effects on N_2O emissions, including suppression [6], stimulation [7,8] and little or no effect. Shakoor et al. [9] conducted a meta-analysis of the worldwide studies on the impact of biochar on mitigating greenhouse gases, showing that biochar application could reduce N_2O emissions in farmland soil. Some studies have demonstrated that biochar changed the abundance of denitrification genes and microbial activities to influence N_2O emissions [10,11]. The application of biochar can improve soil pH, enhance the function of *nosZ* and promote the conversion of N_2O to N_2 in denitrification [12]. Other studies reported that biochar reduced soil NH_4^+-N and NO_3^--N content through its adsorption capacity and high cation exchange capacity, restricting the substrates of nitrification and denitrification, thereby reducing N_2O emissions [13]. However, some experiments' results supported the opposite idea that adding biochar to the soil would enhance N_2O emissions or have no significance effect. There are many influencing factors causing these different results, such as the amount and properties of biochar, soil and climate types, etc. [14,15], which influenced the production process of N_2O, leading to discrepant results. Additionally, with the experiment time extended, the properties of biochar changed [16–18], influencing the N cycle. As a result, no uniform conclusion has been reached about the effect of consecutive biochar application on cropland N_2O emissions.

Similar to the application of biochar, returning straw to cropland, as a direct reuse method of agricultural waste, also had beneficial effects on the soil and affected N_2O emissions [19,20]. Some studies found that straw returns suppressed N_2O emissions [21,22], while others proposed the opposite views [23–25]. Gao et al. [26] conducted a meta-analysis of soil N_2O emissions in wheat–maize rotation systems verifying that there were still different conclusions about the effect of straw return on N_2O emissions. Zhou et al. [27] found that its reduction effect depended on straw decomposition, which accelerates the consumption of soil oxygen and thus promoted the reduction of N_2O during denitrification. While in another study, straw return increased the content of dissolved N and soil organic matter, providing a suitable environment and substrate for microorganisms to perform nitrification and denitrification, and prompting the emission of N_2O [28]. The research on the effect of straw return on N_2O emissions has also drawn different conclusions due to various factors [29], such as the different experimental sites [10,30], straw return methods [31,32], the amount of returned straw [33] and so on. Due to the fact that straw returning to the soil takes a period of time to decompose, the effects of long-term straw return on soil N_2O emissions may vary depending on the duration of the experiment. Therefore, it is necessary to observe N_2O emissions under continuous straw return conditions to clearly understand the long-term effects.

For many years, the extensive application of N fertilizer in North China has led to severe N loss in agricultural ecosystems and increased N_2O emissions from farmlands [34]. Wheat straw and maize straw resources are abundant in North China, accounting for about 25% of straw resources in China, and returning straw to the field is one of the main ways of straw utilization in this area [35]. Based on current research literature, there are few studies to explore the long-term effects of the successive application of biochar and straw return on N_2O emissions. Therefore, it is difficult to assess the long-term impact of biochar and straw return on the environmental and economic benefits. For further research, we carried out 3 years' observation based on the field experiment of continuous application of biochar and straw after 10 years' application in the wheat–maize rotation system in North China. We hypothesized that the successive application of biochar would decrease N_2O emissions compared with straw return and the application of biochar combined with straw. Our objectives are (1) to illustrate the effects of consecutive application of biochar, straw and biochar combined with straw on soil N_2O emissions and crop yield, and evaluate their environmental and economic benefits; and (2) to explore the main driving factors affecting soil N_2O emissions in different growing seasons and years.

2. Materials and Methods

2.1. Experimental Site

The experimental site was located at the North China Intensive Agroecosystem Experimental Station of China Agricultural University in Huantai County, Zibo City, Shandong Province (36°57′ N, 117°58′ E). The area has a temperate monsoon climate. The annual mean precipitation is approximately 560 mm, and the annual mean temperature is 12.5 °C. Field crops are planted in a rotation system with winter wheat (early October to early June of the following year) and summer maize (mid-June to mid-September). The soil has a loam texture with a pH of 8.0 and a bulk density of 1.35 $g \cdot cm^{-3}$. Soil organic carbon content was 12.4 $g \cdot kg^{-1}$, alkali hydrolyzed nitrogen content was 62.4 $mg \cdot kg^{-1}$, available phosphorus content was 21.6 $mg \cdot kg^{-1}$, and available potassium content was 294.0 $mg \cdot kg^{-1}$. The monthly mean temperature and precipitation of the 3 rotation years from 2017 to 2020 in the study area are shown in Figure 1.

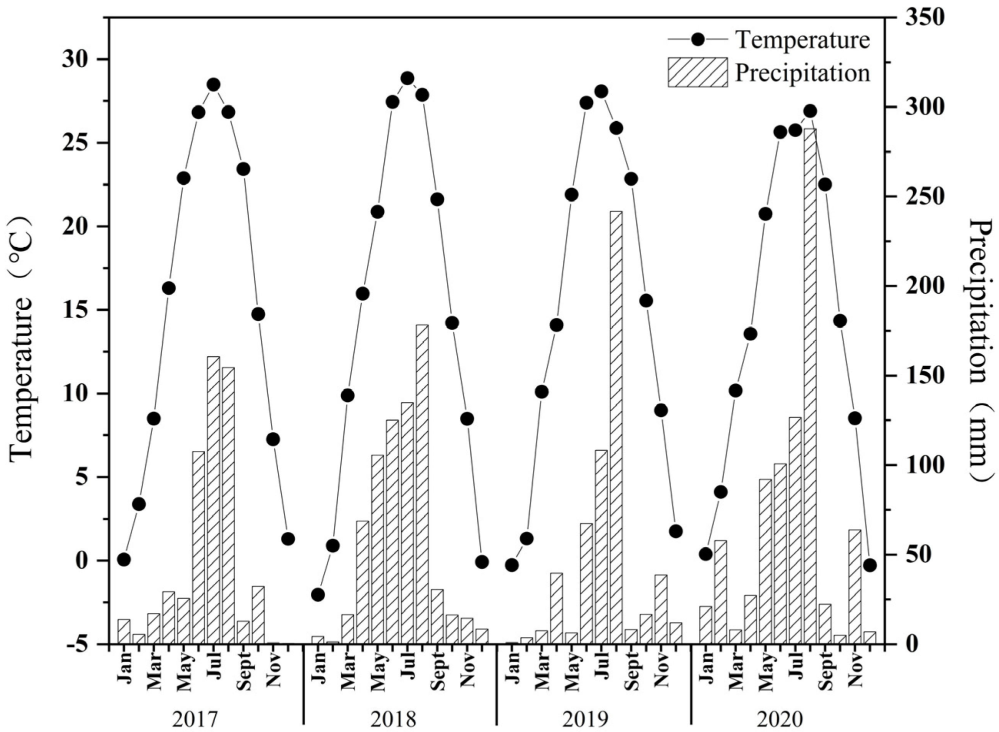

Figure 1. The monthly average air temperature and precipitation from 2017 to 2020 in the study area.

2.2. Experimental Design

The field experiment was started in October 2017 and ended in October 2020, which is 3 complete rotation years. It should be noted that it is based on the long-term positioning experiment that originated in 2007. The field experiment was randomly arranged and each plot covered an area of 6 m × 6 m. There were four treatments (with three replicates): (1) fertilizer (F), the field management only included NPK fertilizer; (2) NPK fertilizer + 9.0 $t \cdot ha^{-1}$ biochar (FB); (3) NPK fertilizer + return of the total wheat/maize straw (FS); and (4) NPK fertilizer + 9.0 $t \cdot ha^{-1}$ biochar combined with the total straw (FSB).

For all treatments, nitrogen (N), phosphorus (P) and potassium (K) fertilizer were applied at rates of 200 kg $N \cdot ha^{-1} \cdot yr^{-1}$, 55 kg $P_2O_5 \cdot ha^{-1} \cdot yr^{-1}$ and 40 kg $K_2O \cdot ha^{-1} \cdot yr^{-1}$. The N content is about 0.5% in the wheat straw, 0.6% in the maize straw, and 0.3% in the biochar, respectively, and the supplementary N fertilizer was replenished by urea in the FB, FS, and FSB treatments. Half of N fertilizer was applied as base fertilizer, and the other half was applied as top-dressing. The biochar used in this study was obtained by pyrolysis of maize straw at 360 °C for 72 h, with a pH of 8.6 and containing 73.0% carbon, 0.325% nitrogen, 0.12% available phosphorus and 1.6% available potassium. Biochar and the base fertilizers were broadcasted onto the soil surface by hand and then immediately incorporated into the soil by tillage to a depth of 15 cm before sowing. For the FS and FSB treatments, all the wheat straw or maize straw produced in the plot was mechanically chopped into 5–10 cm pieces and incorporated into the soil by rotary tillage with the base fertilizer in the following growth season. The C/N ratios of wheat and maize straw were 95:1 and 76:1, respectively.

2.3. Gas and Soil Sampling

2.3.1. Gas Sampling

Gas samples were collected by static chamber method, which consists of a top chamber (45 cm × 45 cm × 50 cm) and a stainless square base placed between ridges. After sowing, a base was placed 20 cm deep into the topsoil and was left in the field for observation throughout the maize or wheat growing season in each plot. The top edge of the base has a groove (5 cm deep) filled with water to seal the edge of the chamber with a horizontal surface. Water was injected into the groove to seal the entire system during sampling.

The top chamber was wrapped with a layer of tinfoil to minimize the changes of the inner air temperature due to sunlight, and it was equipped with a circulating fan inside the top of the chamber to ensure that the gas was completely mixed. Gases were sampled daily for about a continuous one week after sowing, topdressing or irrigation, and once a week at other times. Gas samples were collected between 09:00 and 11:00 local time. Four gas samples were collected at 0, 8, 16, and 24 min with a three-way stopcock using a 50 mL airtight syringe in each plot, and the samples were then injected into pre-evacuated gas bags for analysis. Additionally, the air temperature of the inner chamber and soil temperature were simultaneously measured during each gas sampling. Air temperature was monitored by a sensor probe (JM624, Jinming Instrument CO., Ltd., Tianjin, China) equipped in the top chamber. Soil temperatures at 5 cm depth were determined using a handheld XGN-1000T digital thermal sensor (Yezhiheng Science and Technology Corporation, Beijing, China).

N_2O concentrations were determined by gas chromatograph (Agilent 7890A, Agilent Tech Nologies Inc., Santa Clara, CA, USA) in 24 h and detected by an electron capture detector (ECD). The carrier gas was argon-methane (5%) at a flow rate of 40 mL min^{-1}, and the column temperature was 40 °C. Compressed air was used as a standard gas with a value of 313 ppbv. N_2O concentrations were determined by comparing the peak areas of the samples with those of reference gases. The N_2O concentration fit a linear response with the sampling time in each chamber ($R^2 > 0.9$).

Equation (1) was used to calculate the N_2O flux:

$$F = \rho \times h \times \frac{dc}{dt} \times \frac{273}{273 + T} \tag{1}$$

where F is the N_2O flux [μg·(m^2·h)$^{-1}$]; ρ is the density of N_2O in the standard condition (1.977 g·L^{-1}); h is the height of the chamber (m); dc/dt is the change rate of N_2O concentration in the chamber (μg·h^{-1}); and T is the average temperature in the chamber (°C).

Equation (2) was used to calculate the cumulative N_2O emissions:

$$M = \Sigma \frac{F_{i+1} + F_i}{2} \times (t_{i+1} - t_i) \times 24 \times 10^{-5} \tag{2}$$

where M is the cumulative N_2O emissions (kg·ha^{-1}); F is the N_2O flux [μg·(m^2·h)$^{-1}$]; i is the number of samplings; $t_{i+1} - t_i$ is the number of days between two samplings.

Equation (3) was used to calculate the emission factor of N_2O:

$$\text{N2O emission factor} = \frac{M(\text{fertilizer}) - M(\text{no fertilizer})}{\text{the amount of N fertilizer application}} \tag{3}$$

where M (fertilizer) and M (no fertilizer) are the cumulative N_2O emissions under the condition of applying N fertilizer and no N fertilizer, respectively.

2.3.2. Soil Sampling

Soil samples of 0–20 cm were collected from five random points in each plot with gas sampling. Samples were mixed, passed through a 2 mm sieve and then divided into two parts equally. One part was air-dried at room temperature for the determination of soil pH, and the other part was stored at 4 °C for the determination of soil water content (SWC), NO_3^--N and NH_4^+-N content. Soil pH was measured with a pH meter (Extech, PH100 ExStick, Camarillo, CA, USA) and the SWC was determined using the weighing method by oven drying at 105 °C for 48 h. Soil NO_3^--N and NH_4^+-N were extracted with 0.01 mol·L^{-1} $CaCl_2$ and then determined by automatic flow injection analyzer (Braun and Lubbe, Norderstedt, Germany).

2.4. Statistical Analysis

All data in this study are presented as the mean of three replicates. Excel 2019 was used to process data about N_2O flux, soil physicochemical properties, air temperature, and precipitation, and Origin 2018 software was used for mapping. One-way analysis of variance (ANOVA) was performed to compare data between different treatments by statistical software SPSS26.0. Correlations among N_2O emissions, soil temperature, soil pH, SWC, NO_3^--N content, NH_4^+-N content, air temperature and precipitation were analyzed by cor function in R studio (version 4.1.1). The N_2O emission factors under different treatments in different growing seasons and years was calculated to compare the interannual differences in N_2O emissions.

3. Results

3.1. Soil Physical and Chemical Properties

The soil pH ranged from 7.32 to 8.29 in the first and second rotation cycle, and it increased sharply in the third rotation cycle with the range of 7.60 to 9.50, which was significantly higher than that of the first two rotation cycles (Figure 2a). In the wheat season, compared with the F treatment (7.79), the FB treatment had the highest value in average pH among the other three treatments. In the maize season, the average pH of the FB, FS and FSB treatments were 0.02, 0.10 and 0.11 units lower than that of the F treatment, respectively. Compared with the F treatment (7.89), the annual average pH of the FB treatment (7.91) increased by 0.02 units, and the FS (7.85) and FSB (7.84) treatments decreased by 0.06 and 0.07 units, respectively.

The SWC fluctuated sharply in the three rotation cycles due to precipitation or irrigation, ranging from 7.3% to 27.7%, and the variation trend was similar in all treatments (Figure 2b). The average SWC in the maize season was higher than that in the wheat season in all treatments. The annual average SWC of each treatment was 17.25% (F); 18.28% (FB); 17.89% (FS); and 17.27% (FSB). There was no significant difference in the annual average SWC between these treatments. In the third rotation cycle, compared with the F treatment, the average SWC of the FS and FSB treatments significantly increased by 12.7% and 15.7%, respectively.

As is shown in Figure 2c, the soil NO_3^--N content ranged from 6.1 to 278.1 mg·kg^{-1}, and it peaked after fertilization or topdressing irrigation. The NO_3^--N content of each treatment fluctuated sharply in wheat seasons, and the trend of the variations were basically the same as those in maize seasons. In comparison to the F treatment, the annual average NO_3^--N content of the FB, FS and FSB treatments decreased by 2.9%, 14.6% and 11.4%, respectively.

For the soil NH_4^+-N content, the range of it was 0.01–33.7 mg·kg^{-1} (Figure 2d). A peak appeared after topdressing and irrigation in maize seasons in the first two rotation cycles, and the soil NH_4^+-N content varied slightly in the third year of the experiment, significantly lower than that of the first two years. Compared with the F treatment, the annual average NH_4^+-N content in the FB and FSB treatments decreased by 2.05% and 2.24%, respectively, while it increased by 2.65% in the FS treatment.

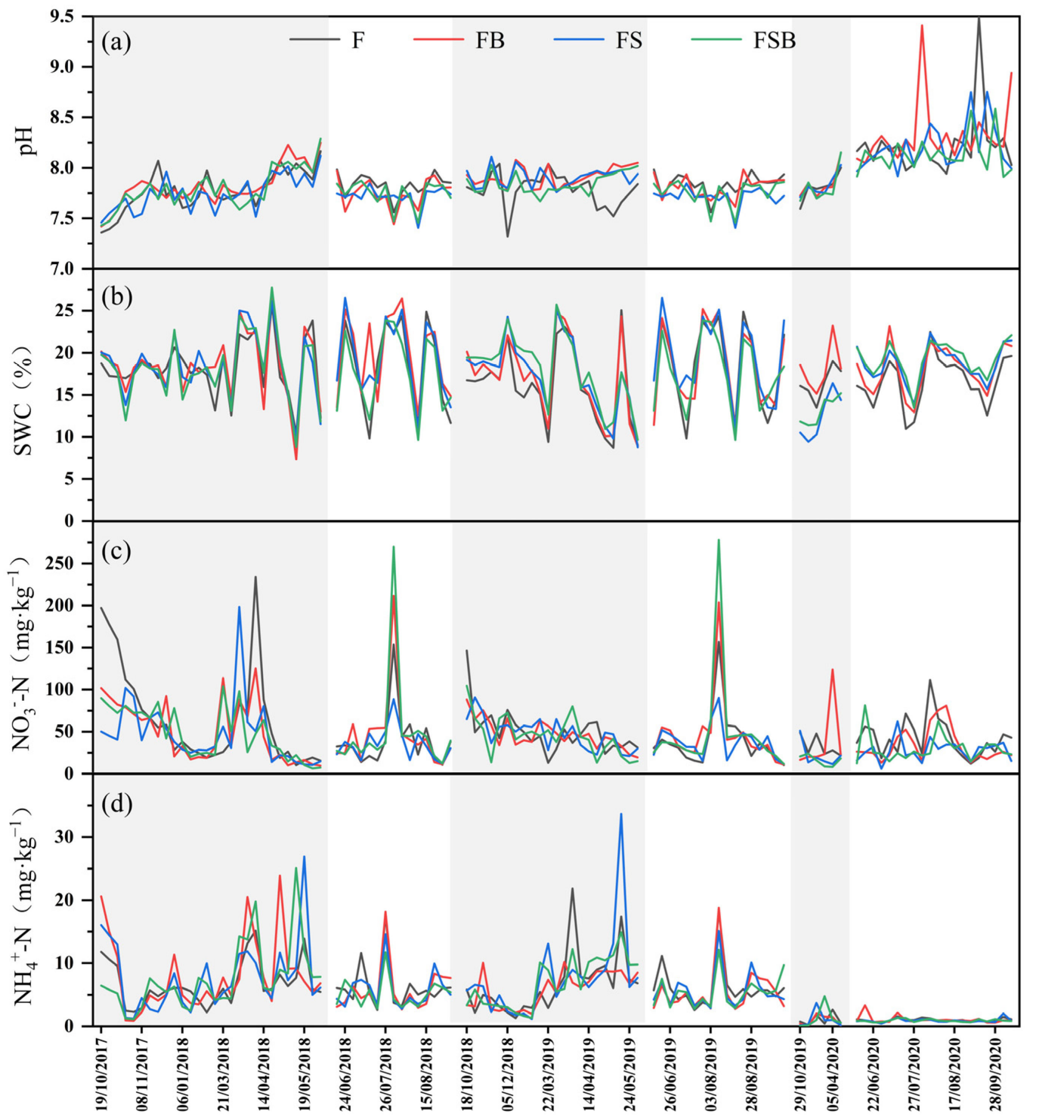

Figure 2. Seasonal variation of soil pH (**a**), SWC (**b**), NO_3^--N content (**c**) and NH_4^+-N content (**d**) from 2017 to 2020 under F, FB, FS and FSB treatments in the experimental site.

3.2. N_2O Emissions

Three years' observations showed that the N_2O flux in the maize season was significantly higher and fluctuated more strongly than that in the wheat season (Figure 3). The cumulative N_2O emissions in the wheat season accounted for 28.0–45.8% of the annual emissions, and 54.2–72.0% of the annual emissions occurred in the maize season. The peak N_2O emission occurred after fertilization or irrigation in all treatments, and the maximum N_2O flux was 2420.6 μg·m^{-2}·h^{-1} in the F treatment in the maize season of 2018, followed by 1972.8 μg·m^{-2}·h^{-1} in the FSB treatment.

In wheat season, compared with the F treatment, the N_2O flux of the FB treatment (46.2 μg·m^{-2}·h^{-1}) decreased by 20.2%, and the N_2O flux of the FS and FSB treatments (78.8 μg·m^2·h^{-1} and 95.1 μg·m^{-2}·h^{-1}) increased by 36.0% and 64.2%, respectively. The variation of N_2O emissions in the maize season was similar to that in wheat season. In the maize season, the N_2O flux of the FB treatment (162.1 μg·m^{-2}·h^{-1}) decreased by 21.7% in comparison with the F treatment, and the N_2O flux of the FS and FSB treatments (218.6 μg·m^{-2}·h^{-1} and 289.3 μg·m^2·h^{-1}) increased by 5.5% and 39.6%, respectively.

For the annual average cumulative N_2O emissions (Figure 4), the lowest emission of 3.10 kg·ha^{-1} was in the FB treatment, and highest emission of 5.50 kg·ha^{-1} was in the FSB treatment. Compared with the F treatment, the FB treatment reduced the cumulative N_2O

emissions by 20.2%, while the FS and FSB treatments stimulated them by 23.7% and 41.4%, respectively. The cumulative N_2O emissions of the FSB treatment in the first rotation cycle were slightly lower than those of the FS treatment, and the cumulative N_2O emissions of the FSB treatment were significantly higher than those of the FS treatment in the other growing seasons. Based on the cumulative N_2O emissions of the three years (Figure 4), the cumulative N_2O emissions of each treatment were, respectively, decreased in the second year, and then increased in the third year. The average N_2O emission flux and cumulative N_2O emissions in the FB treatment were lower than those in the F treatment ($p < 0.05$), while those in the FS and FSB treatments were higher than the F treatment ($p < 0.05$).

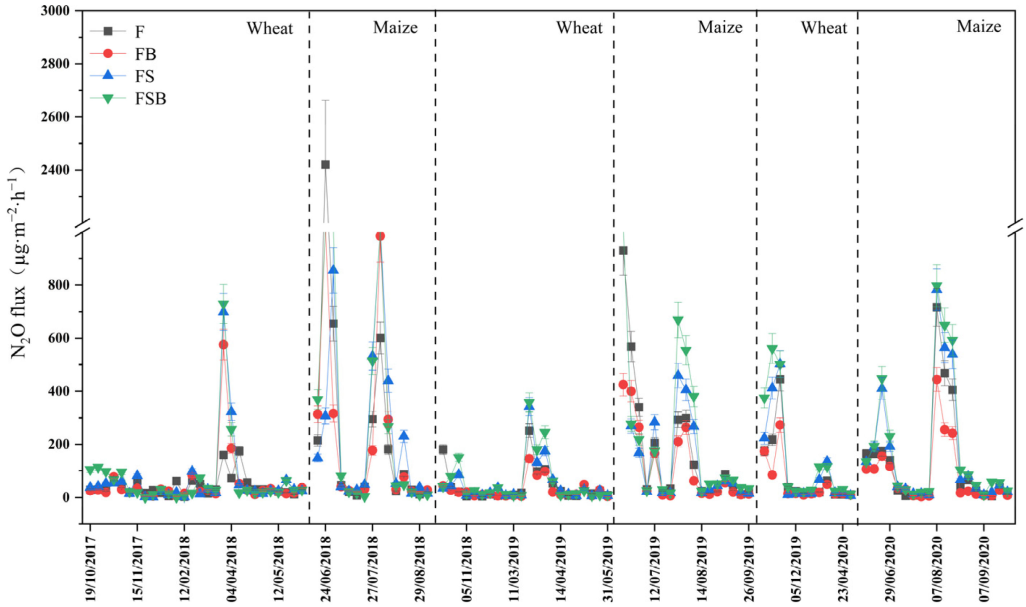

Figure 3. Seasonal variation of N_2O fluxes under F, FB, FS and FSB treatments from 2017 to 2020 in the experimental site.

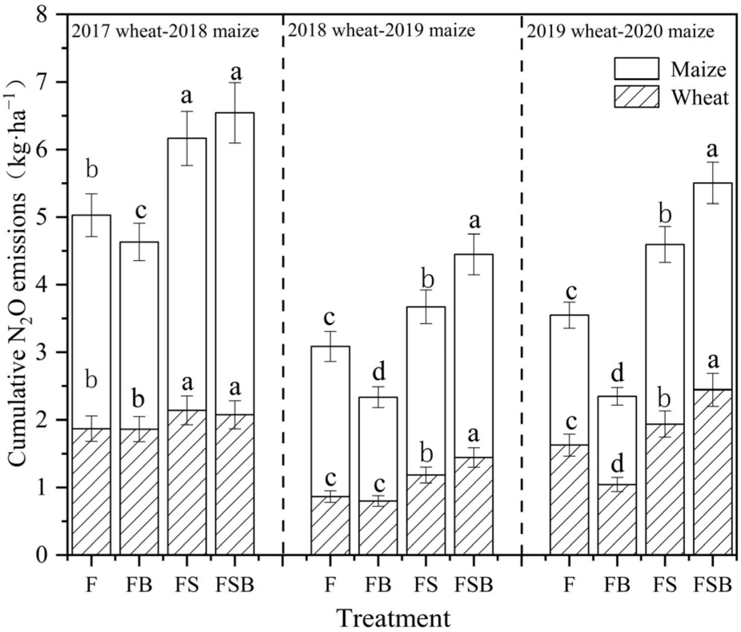

Figure 4. The cumulative N_2O emissions of F, FB, FS and FSB treatments in 2017–2018, 2018–2019 and 2019–2020 rotation cycle. Different letters in the figure indicate significant differences of different treatments.

3.3. N_2O Emission Factor

The result of the soil N_2O emission factor was illustrated in Table 1. In the three rotation cycles, the minimum and maximum N_2O emission factors were 0.47% in the FB treatment and 2.57% in the FSB treatment, respectively. As the experiment went on, the N_2O emission factor of all treatments decreased firstly and then increased. Compared with the F treatment (with an emission factor of 1.24%), the FB treatment reduced the average N_2O emission factor by 32.5%, while the FS and FSB treatments increased them, and the N_2O emission factor of the FSB treatment was 74.7% higher than that of the FS treatment.

Table 1. N_2O emission factors of F, FB, FS and FSB treatment from 2017 to 2020.

Treatment	2017–2018	2018–2019	2019–2020
F	1.81%	0.84%	1.07%
FB	1.62%	0.467%	0.474%
FS	2.38%	1.14%	1.60%
FSB	2.57%	1.52%	2.05%

3.4. Correlations between N_2O Emissions and Environmental Factors

The correlations between N_2O fluxes in each treatment and environmental factors were shown in Figure 5. In the wheat season, the N_2O flux was positively correlated with SWC and NO_3^--N content in the F, FS, and FSB treatment ($p < 0.05$), and it was in highly significant correlation with NO_3^--N content in the FS treatment ($p < 0.01$). N_2O flux had a negative relationship with soil pH in the FSB treatment ($p < 0.05$). In the maize season, the N_2O flux was positively correlated with SWC in the FS and FSB treatments ($p < 0.05$), and it was positively correlated with NO_3^--N content in the F and FB treatments ($p < 0.05$).

3.5. Crop Yield

The wheat yield of each year ranged from 3.61 to 7.55 $t{\cdot}ha^{-1}$ and the maize yield ranged from 6.93 to 8.50 $t{\cdot}ha^{-1}$ (Figure 6), while it was not successively increased year by year. Compared with the F treatment (12.8 $t{\cdot}ha^{-1}$), the FB, FS and FSB treatments increased the annual average yield by 4.99%, 8.40% and10.25%, respectively. There was no significant difference in the yield of each treatment in each season ($p > 0.05$). However, the results about ANOVA analysis of yield-scaled N_2O emissions (Figure 7) showed that the FB, FS and FSB treatments had significant effects on the yield-scaled N_2O emissions ($p < 0.05$). The yield-scaled N_2O emissions of the FB treatment were the lowest, and yield-scaled N_2O emissions of the FSB treatment were dramatically higher than that of the FB treatment in all the growing seasons except the wheat season of 2017–2018 ($p < 0.05$). Different from the trend of the yield, yield-scaled N_2O emissions of the second year were lower than that of the first year. Compared with the F treatment (0.32 $kg{\cdot}t^{-1}$), the yield-scaled N_2O emissions of the FB treatment reduced by 16.4%, and that of the FS and FSB treatments increased by 10.3% and 26.9%, respectively.

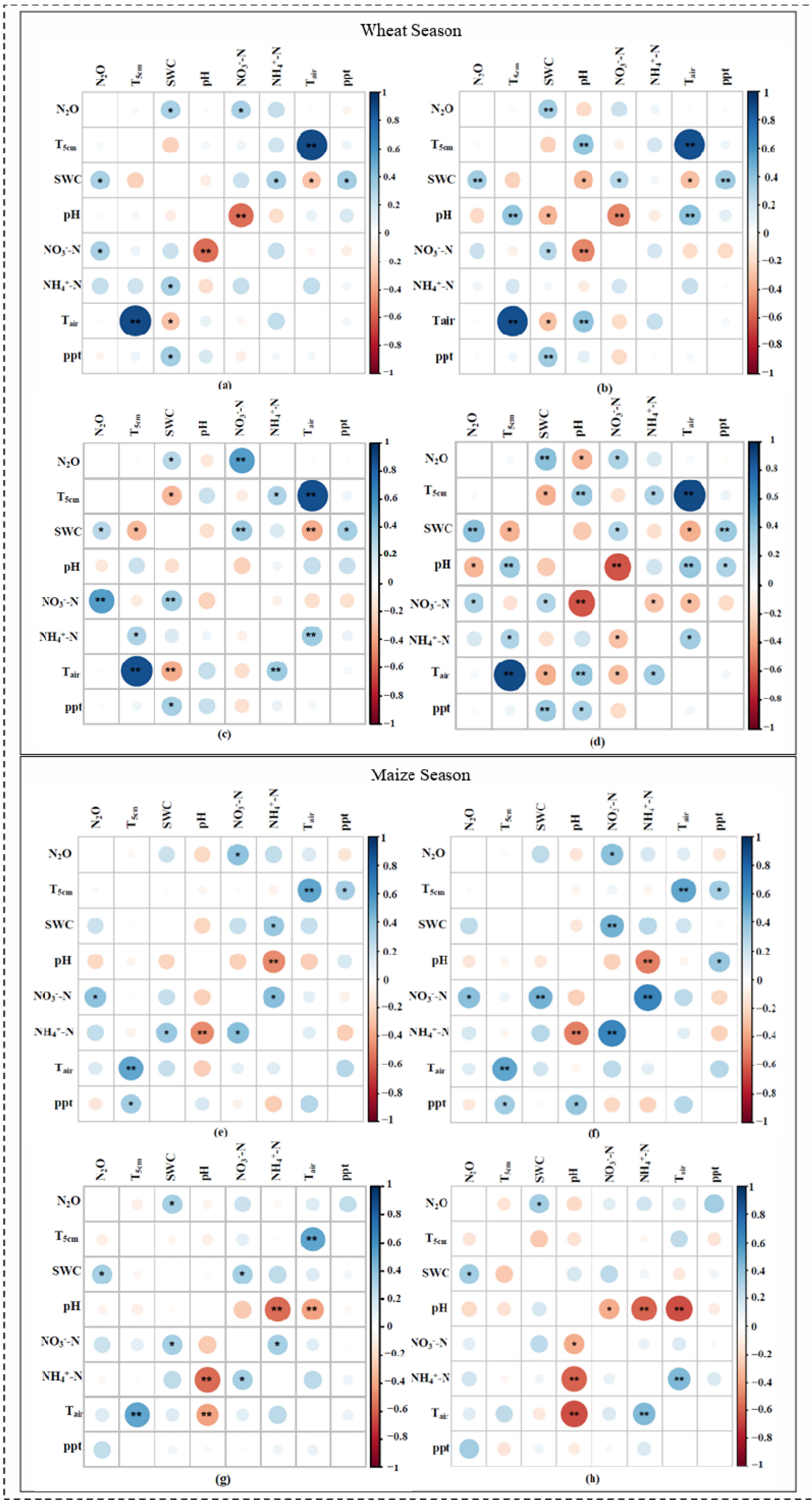

Figure 5. Correlations among N_2O emissions, soil temperature (T_{5cm}), soil pH, SWC, NO_3^--N content, NH_4^+-N content, air temperature (T_{air}) and precipitation (ppt) in different treatments in the wheat and maize seasons. (**a**–**d**) represent the correlations in the F, FB, FS, FSB treatments in the wheat season, respectively; (**e**–**h**) represent the correlations in the F, FB, FS, FSB treatments in the maize season, respectively. ** represents the correlation was significant at level 0.01; * represents the correlation was significant at level 0.05.

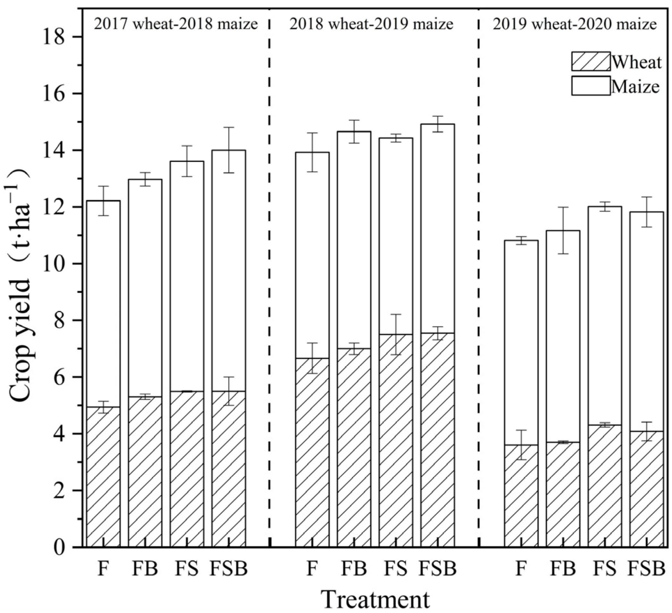

Figure 6. Crop yields of F, FB, FS and FSB treatments in 2017–2018, 2018–2019 and 2019–2020 rotation cycle in the experimental site.

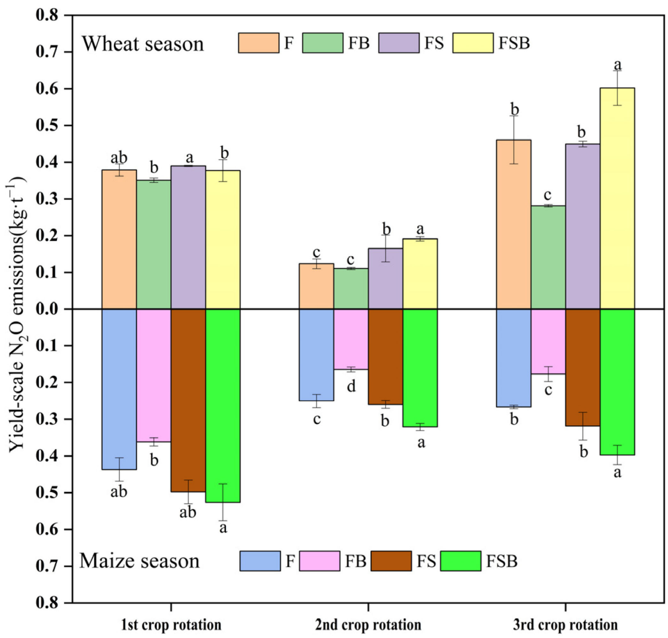

Figure 7. Yield-scale N_2O emissions of F, FB, FS, FSB treatment in different seasons and years from 2017 to 2020. Different letters in the figure indicate significant differences of different treatments.

4. Discussion

4.1. Effects of Environmental Factors on N_2O Emissions

Our results indicated that biochar amendment significantly reduced N_2O emissions by 20.2% (Figure 4), and many relevant studies reached the similar conclusion [36–38]. Dawar et al. [39] found that biochar reduced N_2O emissions by 20–24%, which is close to our results. Additionally, we found that with the continuous application of biochar over the 3-year experiment, the emission reduction effect of N_2O became obvious (Figure 4). However, both straw return and straw combined with biochar significantly increased N_2O emissions by 23.7% and 41.4%, respectively (Figure 4). Similar conclusions were also reached in previous studies [19,30,40]. The main reason was that biochar could immobilize soil NO_3^--N and NH_4^+-N, inhibiting nitrification and denitrification [38,41]. In this study, biochar amendment reduced soil NO_3^--N content (Figure 2), indicating that soil NO_3^--N was fixed by biochar, thereby inhibiting N_2O emissions. For the straw return treatment, some studies have shown that the decomposition of crop straw in soil increases the NO_3^--N content, improves the availability of C and N substrates, and provides suitable substrates for microbial nitrification and denitrification [42,43]. In our study, the FS and FSB treatments reduced NO_3^--N content (Figure 2c), and it may be because more NO_3^--N was involved in denitrification as a substrate and converted into N_2O emissions into the atmosphere. Meanwhile, it has been reported that straw return also promoted the growth of soil microorganisms, enhanced soil respiration, prompted the formation of an anaerobic environment, and thus it accelerated the denitrification rate [10].

In addition, our research found that biochar application increased soil pH (Figure 2). The increased pH in the FB treatment enhanced the activity of nitrous oxide reductase and promoted the reduction from N_2O to N_2 [44,45]. Moreover, biochar played an important role in transferring electrons to denitrifying microorganisms, promoting the conversion of N_2O to N_2, and thus reducing N_2O emissions [46]. Other studies believed that biochar inhibited soil N_2O emissions mainly because it increased the abundance of the soil microbial *nosZ* gene, accelerating the reduction of N_2O [47].

Straw return combined with biochar did not offset the increase in N_2O emissions caused by straw return, and even exceeded the annual average cumulative N_2O emissions of straw return alone. It was found that the soil pH of both straw return and straw return combined with biochar treatments decreased, while the soil pH of biochar treatment increased in this study (Figure 2a). This demonstrated that straw was the main factor influencing N_2O emissions in straw return combined with biochar treatment [48]. On the one hand, the two treatments reduced soil pH (Figure 2a) and inhibited the activity of nitrous oxide reductase, which hindered the process of N_2O conversion to N_2. On the other hand, both treatments increased SWC in the 3rd rotation year (Figure 2b), which easily formed an anaerobic environment in the soil, promoted soil denitrification [27], and stimulated N_2O emissions. Additionally, N_2O emissions were influenced by the C/N ratio. In this experiment, the C/N of biochar was 243:1, and the higher C/N of it broke the original C and N balance in soil, and therefore more N was fixed. This led to the reduction of N substrates for soil denitrification, thus reducing the emissions of N_2O. The C/N ratio of straw was significantly smaller than that of biochar and disturbed the balance between C and N. Thus, straw return can provide enough N to meet the needs of crop growth, improve soil microbial community, and further stimulate N_2O emissions [42], which may also explain the sharp increase in N_2O emissions caused by straw.

Contrary to the results of this study, some studies have shown that the application of biochar has little impact on soil N_2O emissions, the reduction effect was transient or it would increase N_2O emissions conversely [7,49]. Pereira et al. [8] observed that biochar reduced N_2O emissions only for a few days in the test. Van Zwieten et al. [50] reported that biochar addition tended to decrease N_2O emissions by denitrification process, so it made a small effect on N_2O emissions when applying biochar under dry experimental conditions [24]. Other studies have concluded that the application of biochar increased N_2O emissions, which might be due to the different types of biochar used in the experiment, pyrolysis temperature of biochar preparation, and the amount of biochar [51]. For example, Escuer-Gatius et al. [52] found that the hay biochar pyrolyzed at 850 °C would stimulate soil N_2O emission. Bai et al. [40] showed that straw return could promote the conversion of N into N_2O and increased N_2O emissions, because the C in the straw stimulated the growth of microorganisms related to N_2O production, accelerating the nitrification rate. However, Yao et al. [22] did the research in the same experimental site, suggesting that straw return reduced soil N_2O emissions, which may due to different temperature and precipitation conditions in different years.

4.2. Long-Term Effects of Continuous Application of Biochar and Straw on N_2O Emissions

In this 3-year experiment, the application of biochar decreased the N_2O emission factor by 32.5%, implying that the continuous application of biochar produced environmental benefits to some extent. Li et al. [53] reached similar conclusions through a 2-year experiment that biochar amendment reduced the N_2O emission factor effectively in an intensified vegetable field. Some studies have shown that continuous application of biochar for many years was conducive to improving soil properties, providing a favorable living environment for soil microorganisms, thereby improving soil nitrification and denitrification and reducing N_2O emissions [54]. Duan et al. [55] showed that the biochar in soil gradually aged after five-year application, and the decrease in pH value may weaken the inhibiting effect of fresh biochar on N_2O emissions. However, some studies demonstrated that the biochar derived from wheat straw and pyrolyzed at 500 °C could still significantly increase the abundance of AOB and *nosZ* within six years, which promoted the conversion of N_2O to N_2 during denitrification, indicating that biochar has long-term effects on soil N cycle [37,56]. The adsorption capacity of aging biochar to NH_4^+-N was enhanced, which suppressed autotrophic nitrification [57,58], and this also explained the significant decrease in cumulative N_2O emissions after biochar application in the third year compared with the F treatment. He et al. [59] also found that long-term application of biochar could reduce the loss of NO_3^--N, causing a large amount of NO_3^--N to remain in the soil, which also meant that the N substrate involved in soil denitrification was reduced. Moreover, some research reported that after 4 years' application of biochar, there was no significant change in N_2O emissions due to the different N content of the biochar used in the experiment [60].

In our study, treatments involving straw all increased the N_2O emission factor in three consecutive years (Table 1). Consistent with our results, Huang et al. [61] pointed out that conventional fertilization combined with straw return increased the N_2O emission factor compared with only fertilization, but changing the ratio of chemical fertilizer and straw may lower the N_2O emission factor. Long-term straw return can maintain the stability of the soil structure, reduce N leaching and gradually narrow the gap of the C/N ratio between straw and soil, providing more N substrates for denitrification [62]. Therefore, the effect of straw return on N_2O emissions may be affected by the length of returning time. Lehtinen et al. [63] found that straw return less than 5 years had a more significant effect on increasing N_2O emissions than 11 to 20 years. Other studies reported that the initial straw addition provided sufficient C and N substrates for the production of N_2O and stimulated N_2O emissions. However, with the increase in returning times, the continuous addition of straw may reduce N_2O emissions due to the gradual accumulation of residual C and an increase in the soil C/N ratio, leading to N immobilization [64]. In this experiment, N fertilizer was applied as basal and topdressing fertilizer to compensate for the reduction of the N substrate, which was consistent with the research results of Wang et al. [65]. All in all, there are still many uncertain factors regarding the impact of straw return and biochar on N_2O emissions. Therefore, further research is needed on the complex mechanisms of straw and biochar affecting N_2O production in the future.

4.3. Effects of Biochar and Straw on Crop Yield

A large number of studies have indicated that crop yield was affected by factors such as tillage methods, fertilizer types, the amount of fertilizer, climate, and soil properties. In this experiment, the FB, FS and FSB treatments resulted in a slight increase in the crop yield, which is consistent with many studies [66–68]. During the three years, crop yields did not increase continuously, which may be because the crop yield was mainly influenced by climate change in different years. An experiment conducted at the same site showed that biochar and straw treatments had no significant impact on wheat and maize yields [69], and two years of short-term testing might be the main reason. Biochar addition can improve soil structure, reduce soil bulk density, increase soil water capacity, and increase crop yield [70], and this is consistent with the results of this study. Nitrogen is the key factor in maintaining crop yield, and the high cation exchange capacity and adsorption capacity of biochar could reduce the loss of N fertilizer in soil [71,72]. In addition, Dawar et al. [39] also reported that biochar reduced soil N_2O emissions, and retained N used by crops. This also indicates the promoting effect of biochar on crop yield.

Straw return could increase the contents of soil total N, available P and available K, directly increasing soil organic matter and improving soil fertility [73,74]; at the same time, crop straw provides a rich carbon source for soil microorganisms, promoting the recycling and release of soil nutrients by microorganisms. Zhang et al. [75] analyzed the effect of straw return on crop yield based on the relevant data from 1980 to 2014 in the same experimental site. They pointed out that straw return not only increased soil organic carbon content, but also long-term straw return could produce good economic and environmental benefits by increasing soil organic carbon and available nutrient content. Zheng et al. [76] also found that returning straw to the field at the same experimental site was beneficial for soil carbon sequestration and increased crop yield. The combined application of biochar and straw could also increase crop yield, as the addition of straw stimulates the decomposition of organic matter by soil microorganisms and biochar absorbed it. The combination of biochar and straw prevented N losing and improved N use efficiency [77].

Apart from that, in terms of the yield-scaled N_2O emissions, the emissions of biochar amendment were significantly lower than that of the FS and FSB treatments (Figure 7), indicating that applying biochar significantly reduced N_2O emissions when producing the same yield. Comprehensively considering the crop yield and the yield-scaled N_2O emissions, only biochar amendment decreased the yield-scaled N_2O emissions (Figure 7), although higher yield appeared in the FS and FSB treatments (Figure 6). This indicated that the application of biochar can maintain crop yield and inhibit N_2O emissions from soil concurrently. All in all, the benefits of straw carbonization returning to the field are much greater than that of straw direct returning to the field. Additionally, the cost of producing biochar is worth noting. Hence, to maximize the function of biochar, studies about optimizing and upgrading the production of biochar should be continued.

5. Conclusions

Three consecutive years of soil physicochemical properties, crop yields and N_2O emissions showed that the long-term application of biochar reduced soil N_2O emissions, while straw return and straw combined with biochar increased N_2O emissions. Soil NO_3^--N content, SWC and soil pH were the main influencing factors affecting N_2O emissions in wheat season, and SWC and NO_3^--N content were the main influencing factors in maize season. In conclusion, continuous application of biochar can significantly reduce N_2O emissions while maintaining similar crop yield. Biochar combined with straw treatments can improve the crops yield, but they are not beneficial to suppressing N_2O emissions. Based on the research results, the 3-year experiment applying biochar following 10 years' addition has demonstrated the long-term beneficial effects of biochar on N_2O emissions reduction. Future research should pay more attention to the mechanism of biochar reducing N_2O emissions, elaborating the main influencing factors of N_2O emission reduction through different microbial pathways, and the impact of different C:N ratios on N_2O emission reduction. It is also important to evaluate the environmental and economic benefits of biochar in a future study.

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