EFFECT OF CH_4 EMISSION REDUCTION OF WATER, FERTILIZER AND BIOCHAR REGULATION METHOD ON A RICE FIELD IN THE NORTHEASTERN COLD AREA OF CHINA

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Abstract. The objective of this study was to analyse the black soil rice fields of the northeastern cold region of China**,** D311 optimal design scheme with three factors secondary saturation was adopted and static opaque chamber - gas chromatographic method was utilized to analyze the effect of irrigation amount, nitrogen fertilizer and straw biochar on the emission of the greenhouse gas CH_4 from rice fields, the study determined the optimal application scheme of water and fertilizer for emission control. The results show that the order of influence for these factors from the highest to the lowest is: biochar > nitrogen fertilizer > water; effect of irrigation amount on CH_4 emission is increased at first, followed by a decrease. Increase of nitrogen fertilizer and biochar can significantly reduce CH_4 emission loads; interaction between two factors has an inhibitory effect on CH_4 emissions and it is shown as below: nitrogen fertilizer + biochar > water + biochar > water + nitrogen fertilizer; in combination with the yield, when emission reduction target of rice field CH_4 is controlled at 20~40% of normal emission, the optimized application scheme in combination of water, fertilizer and biochar is the following: irrigation amount 4,930-5,310 m^3/hm^2, nitrogen application amount 96.93-107.74 kg/hm^2 and biochar application amount 19.71-24.12 t/hm^2.
Keywords: *cold black soil, rice, irrigation and fertilization, biochar dosage, CH_4 emission*

Introduction

The reason for global warming lies in the increase of greenhouse gas concentration in the atmosphere. CH_4 as an important greenhouse gas in the atmosphere has already contributed to as much as 15% for greenhouse effect. Hence, CH_4 has become a key factor affecting global climate following CO_2 (Wang, 2001; Jiang, 2001; Wang et al., 2008). Hence, rice fields are main anthropogenic source for CH_4 emission. About 3.1×10^{10}~1.12×10^{11} kg released annually from rice fields accounts for 5~19% proportion in CH_4 emission load to the atmosphere (Zou et al., 2009; IPCC, 2007).

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Therefore, the reduction of greenhouse gas CH_4 emission from rice fields has a great significance in mitigating climate change in China.

There are many factors affecting CH_4 emission from rice fields. It is shown from the study that irrigation mode of rice fields plays an important role in CH_4 emission. Compared with submerged irrigation, CH_4 emission from rice fields under inadequate irrigation will be significantly reduced (Peng et al., 2010; Li et al., 2005). Similarly, fertilization measures have an important impact on CH_4 emission from rice fields. It is shown from the study that the application of fertilizer can increase $NH_4{}^+$-N concentration, but growth of CH_4 oxidizing bacteria is also promoted. The promoted CH_4 oxidizing bacteria oxidize more CH_4, which results in the reduction of CH_4 emission from rice fields (Zou et al., 2005; Cai et al., 1997; Ma et al., 2007). Biochar can play an important role in the global carbon geochemical cycle, climate change and environmental system because of its strong ability of nitrogen and carbon fixation. It has become a hot topic in atmospheric science and environmental science fields (Liu, 2011.). The study shows that applying biochar to soil can significantly improve soil quality and permeability. While fixing atmospheric CO_2, soil CH_4 emission (Karhua et al., 2011; Qin et al., 2012) was also affected. Indoor pot experiments performed by Rondon showed that CH_4 emission decreased by 20.4% (Rondon et al., 2007) when 2 kg/m^2 biochar was added to the soil for cultivation of forage and soybean.

At present, although many scholars have conducted in-depth studies on the effects of water, fertilizer and biochar management on CH_4 emissions from rice fields (Shi et al., 2011; Yuan et al., 2008; Liang et al., 2004; Knoblauch et al., 2008; Xu et al., 2015), they are basically individual single-factor studies, but the effects of integrated management on CH_4 emissions from rice fields (factor coupling effect) are rarely reported. The objective of this study was mainly to analyze the coupling effects of three factors water, nitrogen fertilizer and biochar on CH_4 emissions from rice fields. In combination with CH_4 emission reduction targets during rice growing reason, optimal water, fertilizer and biochar application schemes are sought so as to provide the field management technology reference for CH_4 emission reduction in black soil rice fields in the cold area of Northeastern China.

Materials and methods

Overview of experimental sites

The experiment was carried out at the Rice Irrigation Test Center Station (125°44’E, 45°63’N) in Heping Town, Qingan County, Suihua City, Heilongjiang Province from May to October, 2018, which is a typical cold black soil area. With 2.5 ℃ annual mean temperature, 550 mm annual mean precipitation, 750 mm annual mean evaporation from water surface, 156~171 d hydrothermal growth period of crops and 128 d frost free period all year round, the area based on the climatic characteristics is classified as continental monsoon climate in cold temperature zone. As rice soil is an albic soil type, the soil here has 1.01 g/cm^3 unit weight and 61.8% porosity. The basic physicochemical properties of the soil are the followings: organic matter mass ratio 41.4 g/kg, pH value 6.40, total nitrogen mass ratio 15.06 g/kg, total phosphorus mass ratio 15.23 g/kg, total potassium mass ratio 20.11 g/kg, alkali hydrolysis nitrogen mass ratio 154.36 mg/kg, available phosphorus mass ratio 25.33 mg/kg and available potassium mass ratio 157.25 mg/kg.

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Experimental design

Saturated D311 optimal design (Xu, 1997) is used in the experiment to study the effects of irrigation amount, nitrogen fertilizer and biochar on CH_4 emission in rice growing season under controlled irrigation conditions. Water and fertilizer are applied based on the application standard of local farmers, namely 2,500~7,500 kg/hm^2 irrigation amount, 50-150 kg/hm^2 nitrogen fertilizer (pure nitrogen) and 0~40 t/hm^2 biochar. The detailed design plan is shown in *Tables 1* and *2.*

Table 1. *Encoding table for factor level*

Encoding value			Practical value		
X_1	X_2	X_3	W (m^3/hm^2)	N (kg/hm^2)	BC (t/hm^2)
2	2	2	7500	150	40
1.414	1.414	1	6800	135	30
0	0	0	5000	100	20
-1.414	-1.414	-1	3200	65	10
-2	-2	-2	2500	50	0

W (X1) - water, N (X2) - nitrogen fertilizer, BC (X3) - biochar

Table 2. *Optimal design treatment table of saturated D-311*

Treatment no.	Encoding value			Practical value			CH_4 emission load (kg/hm^2)
	X_1	X_2	X_3	W (m^3/hm^2)	N (kg/hm^2)	BC (t/hm^2)	
1	0	0	2	5000	100	40	121.29
2	0	0	-2	5000	100	0	209.71
3	-1.414	-1.414	1	3200	65	30	179.42
4	1.414	-1.414	1	6800	65	30	179.95
5	-1.414	1.414	1	3200	135	30	158.29
6	1.414	1.414	1	6800	135	30	150.45
7	2	0	-1	7500	150	10	164.63
8	-2	0	-1	2500	50	10	147.99
9	0	2	-1	5000	100	10	165.49
10	0	-2	-1	5000	100	10	178.17
11	0	0	0	5000	100	10	144.42

W (X1) - water, N (X2) - nitrogen fertilizer, BC (X3) - biochar

Eleven treatments with three repetitions are arranged in randomized block. Each block covers a 10 m*10 m = 100 m^2 area. Around the block, rice was also planted so as to add the protection line. With the same rice seedling raising, transplanting, plant protection, medication and other technical measures as well as field management conditions, the blocks were separated with impervious treatment measures to decrease the effect of lateral infiltration on the test, namely plastic sheets and cement ridges were used as seepage isolation materials around the blocks. They were buried 40 cm deep into the surface of the field. Pipeline water supply was adopted. Each pipeline was equipped with water meters so as to control the irrigation amount. Nitrogen fertilizer was applied according in 5:3:2 ratio of base fertilizer, tillering fertilizer and spike

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fertilizer. P fertilizer used as base fertilizer was applied at a time with 45 kg/hm^2 application amount. K fertilizer was applied twice as base fertilizer and 8.5 leaf age (panicle primordium differentiation stage) with 1:1 ratio. With 80 kg/hm^2 application amount, biochar was applied to the surface of the soil and then evenly mixed with plowing soil by rotary tiller. The tested fertilizers are urea (containing N 46%), diammonium phosphate (containing N18%, containing P_2O_5 46%) and potassium fertilizer (containing 40% K_2O). The tested biochar is the s rice traw biochar product supplied by Liaoning Golden Future Agriculture Technology Co., Ltd. The physical and chemical data are shown in *Table 3*.

Table 3. *Physicochemical data of rice straw biochar*

	PH (H2O)	C (%)	N (%)	P (%)	K (%)	CEC (cmol.kg^{-1})	Surface area (m^2·g^{-1})	Void area (cm^3.g^{-1})
Rice traw biochar	10.2	42.7	0.76	0.16	1.07	44.7	81.8	0.08

The rice varieties tested were Longqing Rice No. 3 with the planting density of 4 plants per hole, 25 holes per square metre. Base fertilizer was applied on 6 May and transplantation was performed on 17 May. Tillering fertilizer was applied on 31 May, earing fertilizer was applied on 19 July and the rice was harvested on 20 September. 127 d growth period of rice was divided into period of seedling establishment (May 17-May 30), tillering period (May 31-July 7), jointing and booting period (July 8-July 25), heading to flowering period (July 26-August 4), milk ripe period (August 5-August 24) and yellow ripening period (August 25-September 20).

Gas collection and determination

Gas sampling and selection were carried out on sunny days by static opaque chamber-gas chromatography method. The box is a cuboid with a cross section of 18 cm side length. It is made of plexiglass. Insulation material (sponge and aluminum foil) is pasted on the outside of the box to reduce the gas temperature change in the box caused by solar radiation during sampling. In the early growth stage, the box was 90 cm high and the box increased to 130 cm high after heading stage. A three-way valve gas recovery hole is on the box side 30 cm from the top connecting the three-way valve and gas collector. One fan is built at top of the sampling box so as to mix gas uniformly in the box during sampling. Before transplanting, a wooden base is placed in the sampling basin and aligned with the mud surface. During gas sampling, the sampling box is gently placed on the base of the concentric-circle-liked frame. The water in the base flume guarantees the gas isolation between the inside and outside of the sampling box during sampling. One week after rice transplantation, detection was started. The detection was performed from 10:00 to 12:00 (Li et al., 1998; Epstein and Burke, 1998). At each treatment, gas was collected for three times in parallel weekly until one week before harvest. About 100 mL gas in the box was extracted with a syringe during sampling. Samples were collected at 0, 5, 10 and 15 min, respectively. Afterwards, the gas in the syringe was transferred to the aluminium foil sampling bag immediately, and the sampling bag was brought back to the laboratory in time for determination.

Gas CH_4 concentration was detected with Shimadzu GC-14B meteorological chromatograph along with hydrogen flame ionization detector (FID) and thermal

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conductivity detector (TCD) at 200 ℃ and 100 ℃ temperature. The separation materials were GDX-502 and Porapak Q, respectively and the column temperature was 100 ℃ and 55 ℃. The standard gas was provided by the National Standard Materials Center. The gas collection device is shown in *Figure 1*.

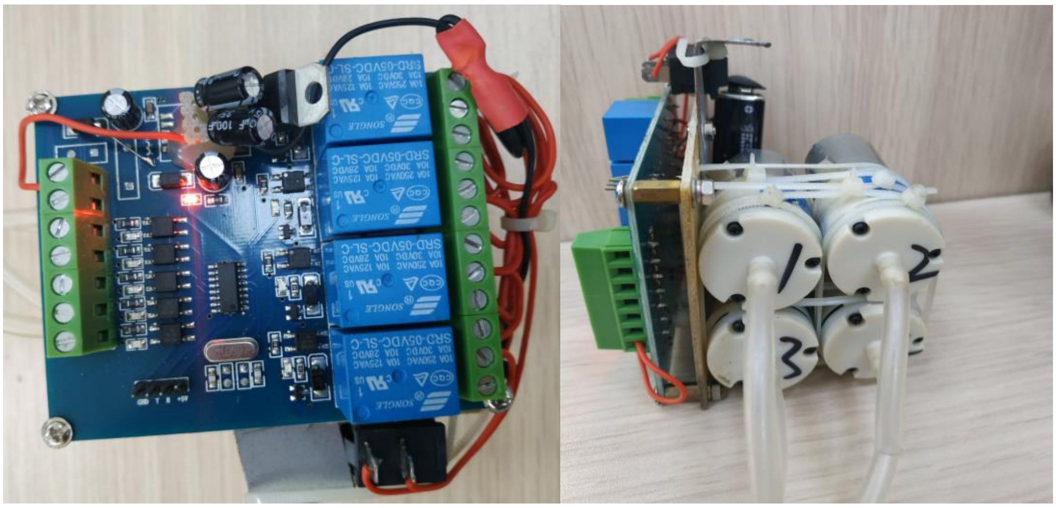

***Figure 1.** A device for automatically collecting static chamber greenhouse gases*

Calculation method and data analysis

The following formula was used to calculate CH_4 emission flux from rice field (Zheng et al., 1998): $F = \rho \cdot h \cdot dc / dt \cdot 273/(273 + T)$, where F is gas emission flux ($mg.m^{-2}.h^{-1}$), ρ is gas density under standard state ($kg.m^{-3}$), h is box height (m), dc/dt is the gas concentration change rate in the sampling box ($mL \cdot m^{-3} \cdot h^{-1}$), 273 is the gas equation constant and T is the average temperature in the sampling box during the sampling process (℃). According to the relationship curve between gas concentration and time, the gas emission flux was calculated. The emission load during the growing season was the accumulated products of average flux value of each growing period and total duration in the growing period (Singh et al., 1996).

The data were analyzed with Excel 2003, SPSS 17.0 and MATLAB 7.0, using regression analysis and variance analysis to process the experimental data.

Results and analysis

Emission load effect function of CH_4 in growing season

The coding values X_1(W), X_2(N) and X_3(C) in *Table 1* were taken as independent variables and the average emission value of CH_4 in the growing season in *Table 2* were taken as the dependent variables for quadratic polynomial regression analysis so as to obtain the regression equation among CH_4 emissions load, irrigation amount, nitrogen fertilizer and biochar.

$$Y = 161.92 + 1.43X_1 - 6.05X_2 - 10.31X_3 - 1.05X_1X_2 - 2.73X_1X_3 - 2.89X_2X_3 - 5.05X_1^2 - 1.21X_2^2 - 0.57X_3^2 \quad \text{(Eq.1)}$$

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F test was performed for the regression equation: F = 5.16 > ($F_{0.01}$(10, 20)) = 3.37). The regression equation has a very significant relationship, that is the equation can reflect the relationship among CH_4 emission load in growing season and irrigation amount, nitrogen fertilizer and biochar. The absolute value of the first term coefficient of the regression equation is the basis for judging the influence degree of each factor on CH_4 emission. Therefore, the influence degree of the equation on CH_4 emission load from high to low was biochar, nitrogen fertilizer and water.

Single factor effect analysis

"Dimension reduction method" was adopted for the above main effect model. Any two factors were fixed at zero code value so as to determine the influencing effect of a single factor on the emission load of CH_4 during the growing season, and then respectively obtain single-factor effect equation and draw single-factor effect curve (*Eq. 2*).

$$Y_1 = 161.92 + 1.43X_1 - 5.05X_1^2 \quad \text{(Eq.2)}$$

$$Y2 = 161.92 - 6.05X_2 - 1.21X_2^2 \quad \text{(Eq.3)}$$

$$Y3 = 161.92 - 10.31X_3 + 0.57X_3^2 \quad \text{(Eq.4)}$$

It can be observed from *Figure 2* that within the coding value range, the effect from irrigation amount on CH_4 emission amount is promoting at first and then restraining. Increase of nitrogenous fertilizer and charcoal can significantly restrain CH_4 emission load.

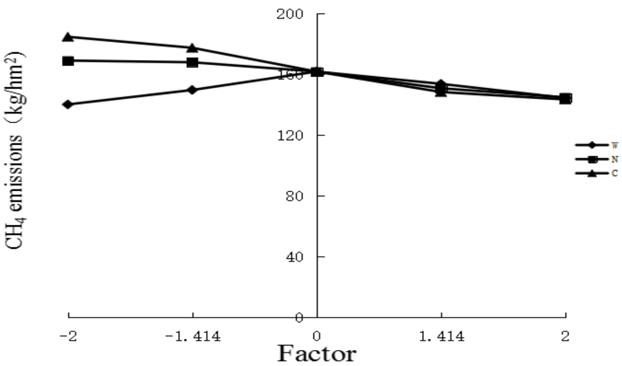

Figure 2. *Single-factor effect curve diagram. (X1 - water, X2 - nitrogen fertilizer, X3 - biochar)*

Interactive effect analysis of factor

Any factor is fixed at zero code value to obtain the interactive effect equation of two other factors and the equation is shown below:

$$Y_{12} = 161.92 + 1.43X_1 - 6.05X_2 - 1.05X_1X_2 - 5.05X_1^2 - 1.21X_2^2 \quad \text{(Eq.5)}$$

$$Y_{13} = 161.92 + 1.43X_1 - 10.31X_3 - 2.73X_1X_3 - 5.05X_1^2 - 0.57X_3^2 \quad \text{(Eq.6)}$$

$$Y_{23} = 161.92 - 6.05X_2 - 10.31X_3 - 2.89X_2X_3 - 1.21X_2^2 - 0.57X_3^2 \quad \text{(Eq.7)}$$

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A diagram is drawn for the interactive effect equation of these two factors (*Fig. 3*). It can be seen from *Figure 3*, the interaction between two factors has an inhibitory effect on CH_4 emission and the effect degree on CH_4 emission load from high to low is as follows: nitrogen fertilizer + biochar, water + biochar and water + nitrogen fertilizer. As can be seen from *Figure 3a* and *b*, when irrigation amount is fixed at a certain level, CH_4 emission load decreases with the increase of the application of nitrogen fertilizer and biochar. However, when nitrogen fertilizer or biochar is fixed at a certain level, the effect of irrigation amount on CH_4 emission increases or decreases and no obvious emission reduction can be obtained; it can be observed from *Figure 3c* that with increase in application quantity of nitrogen fertilizer and biochar, CH_4 emission decreased significantly. Thus, increase of biochar application quantity has a significant effect on CH_4 emission reduction effect.

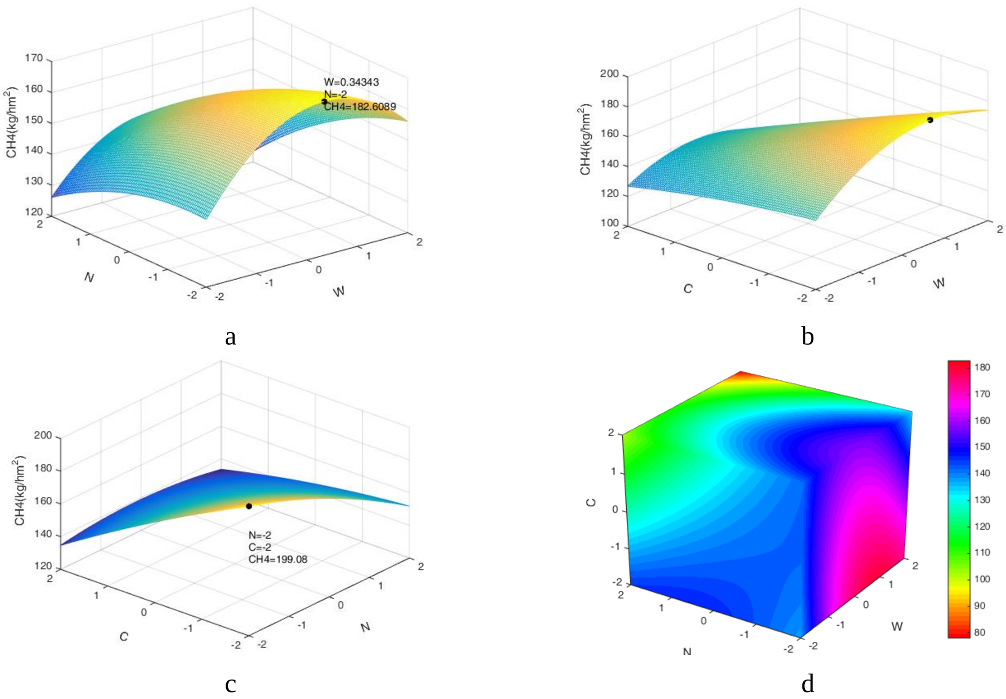

***Figure 3.** Interactive effect analysis between two factors related to emission load of methane in growing season. Interactive effect curve diagram between (a) irrigation amount and nitrogenous fertilizer, (b) irrigation amount and biochar and (c) nitrogenous fertilizer and biochar. (d) Interactive effect four-dimension diagram among irrigation amount, nitrogenous fertilizer and biochar. (W - water, N - nitrogen fertilizer, C - biochar)*

Analysis for management and optimization plan of water, fertilizer and biochar

Frequency analysis method was used to optimize the main effect model. The coding values were divided into five levels (-2, -1.414, 0, 1.414, 2) within the experimental design range to constitute $T = 5^3 = 125$ treatment combinations. Combining with the yield, the emission reduction target of CH_4 in rice fields during the growing season was controlled within 20-40%, because all the factors of treatment No. 11 are at zero level in this experiment, so they are regarded as normal treatment, that is, 60~80% (86.65~115.54 kg/hm^2) normal emission load of CH_4 in growing season was selected for frequency analysis to obtain 50 optimization results for the management simulation

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equation between water fertilizer and biochar in CH_4 emission load during the growing season. Frequency analysis of gas emission flux is shown in *Table 4*.

Table 4. *Water and fertilizer and biochar application plan with CH_4 emission of 86.65~115.54 kg/hm^2 in the rice growing season*

Coding value	Irrigation amount		Nitrogenous fertilizer		Biochar	
	Times	**Frequency/%**	**Times**	**Frequency/%**	**Times**	**Frequency/%**
-2	9	18	9	18	4	8
-1.414	9	18	10	20	12	24
0	9	18	9	18	21	42
1.414	10	20	10	20	8	16
2	13	27	12	24	5	10
Average value	0.19		0.08		0.09	
	0.11		0.11		0.08	
Standard error confidence interval (95%)	-0.0564~0.2480		-0.1230~0.3094		-0.0287~0.4124	
Optimal plan (kg/hm^2)	4930~5310		96.93~107.74		19.71~24.12	

Discussion

Irrigation amount has an important influence on CH_4 emission load from rice fields. It is shown from the study that relatively small irrigation amount can promote gas exchange between soil and atmosphere, destroy the anaerobic conditions of soil and inhibit the activity for production of CH_4 bacteria. However, increase of soil aeration promotes CH_4 emission directly into the atmosphere to a certain extent. Whereas, in case of sufficient water quantities, the rice field will keep a deep water layer for long time. The air and soil is blocked by water layer, which may close some stomata and reduce CH_4 emissions via plants (Ding, 1997; National Information Bulletin on Climate Change of the People’s Republic of China, 2004). This is consistent with the study results.

Nitrogen fertilizer application amount CH_4 has an important influence on CH_4 emission from rice field. The experimental results showed that the nitrogen fertilizer applied has obviously a negative effect on CH_4 emission load from black soil rice field in the growing season, which is basically consistent with the results of Shangguan et al. (1996) and others think that urea could reduce CH_4 emission of rice fields. However, Liang et al. (2002) and others believe that urea has different effects on CH_4 release (promoting or inhibiting), possibly because it can increase the soil pH value. In most cases,When urea was applied to acid soil, the increase of pH value of soil became favorable to the formation of CH_4, Whereas, most black soils are neutral and alkaline. After urea is applied to neutral and alkaline soils, increased pH value restrains formation of CH_4. Therefore, the effect of nitrogen fertilizer application on CH_4 emission of rice fields requires to be further studied.

The results show that biochar application can effectively reduce CH_4 emission load of rice fields. The reason may be that input of biochar effectively improves soil aeration, reduces soil water-soluble organic carbon content, thus improving soil fertility. In addition, biochar input as a carbon source can provide sufficient matrix for CH_4 oxidizing bacteria and reduce CH_4 emissions via oxidation (Liang et al., 2002).

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Conclusion

(1) Water, nitrogen and biochar have different degree of effects on CH_4 emission load of rice fields during growing season. The analysis results show that the effects of three factors on CH_4 emission load are as follows: biochar > nitrogen fertilizer > water; effects of irrigation amount on CH_4 emissions are increase at first and then decrease. Increase of nitrogen fertilizer and biochar can significantly reduce CH_4 emission load.

(2) The interaction of two factors can inhibit CH_4 emission during growing season. The results show that the effects on CH_4 emissions are as follows: nitrogen fertilizer + biochar > water + biochar > water + nitrogen fertilizer; when irrigation amount is fixed at a certain level, CH_4 emission amount decreases with the increase of application amount of nitrogen fertilizer and biochar. However, when nitrogen fertilizer or biochar is fixed at certain level, the increase of irrigation amount may cause CH_4 emission load increase or decrease. There is no obvious emission reduction effect. With increase of application amount of nitrogen fertilizer and biochar, CH_4 emission amount decreased significantly. Thus, increase in application amount of biochar has an obvious effect on CH_4 emission reduction.

(3) In combination with the yield, it is to reduce CH_4 emission load of rice fields in the growing season is controlled from 20 to 40%. The frequency analysis method is used to optimize the main effect model. The optimized combined application plan of water, fertilizer and biochar was determined as follows: irrigation amount 4,930~5,310 m^3/hm^2, nitrogen application amount 96.93~107.74 kg/hm^2 and biochar quantity 19.71~24.12 $t/hm^{2.}$

In this study, we tried to add biochar into the soil to realize joint coupling with water and fertilizer, and established a mathematical model of CH_4 emission load during the growing season of rice fields in the Northeast of China about water, nitrogen and biochar. The model can reflect the relationship between CH_4 emission from rice field and water, fertilizer and biochar through significance test, so as to make quantitative research on water, fertilizer and biochar more convenient. Therefore, the research has a good application prospect. However, the coupling effects of water, fertilizer and biochar on seasonal CH_4 emissions of rice fields have only been preliminarily discussed in this paper, but no qualitative research has been carried out. In the process of further in-depth study, data of different growth stages for many years should be accumulated to make the model more perfect with practical guiding significance.

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