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ORIGINAL ARTICLE

Contrasting emissions of carbon-based greenhouse gases from two paddy soils under submerged conditions as affected by biochar addition

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Abstract

Because two carbon-based greenhouse gases, CH_4 and CO_2, can share a common soil carbon source, reduced emissions of one gas may enhance emissions of the other. The current study aimed to compare CH_4 and CO_2 emissions and identify some related mechanisms caused by biochar addition to paddy soils. Two soils of relatively high organic carbon (OC = 3.05%) and low OC (OC = 0.54%) were taken from two paddy fields, mixed with biochar at 0%, 2%, and 4%, submerged, and incubated in closed plastic jars for 53 days. On days 1, 4, 8, 13, 19, 26, 34, 43, and 53, gas from the jar's headspace was measured for CH_4 and CO_2, and standing water in individual jars was measured for pH and EC. The results revealed that biochar reduced CH_4 emissions by 13% and 74% while increasing CO_2 emissions by 36% and 86% in high-OC and low-OC soil, respectively. Biochar had greater impacts on pH, EC, and greenhouse gas emissions in low-OC soil than in high-OC soil. The inverse relationship between CH_4 emissions and pH and EC can be explained by biotic mechanisms, which suppressed microbial activities, lowing CH_4 emissions. CO_2 emissions were proportionally or non-significantly correlated with EC and pH depending on tested soils, which may be explained by abiotic processes. In summary, biochar addition can suppress CH_4 emissions while increasing CO_2 emissions via biotic and abiotic mechanisms, respectively, dependent on soil pH and EC changes, and biochar effects were stronger in low-OC soil than in high-OC soil.

Keywords Biochar · CO_2 emissions · Methane emission · Paddy soils · Submerged conditions

Introduction

Carbon dioxide (CO_2) and methane (CH_4) are the two important anthropogenic carbon-based greenhouse gases (GHG), contributing to the present trend of global warming (IPCC 2019). The two GHGs can be produced from decomposed fractions of organic carbon (OC) of paddy soil with varying soil organic carbon (SOC) contents. To increase soil productivity, biochar (BC) has been used as a soil amendment, which brings a lot of benefits to rice crop growth (Jeffery et al. 2011; Lu et al. 2020; Mohammadi et al. 2020). Nonetheless, biochar is a carbon-rich substance having some considerable proportion of decomposable carbon (Calvelo Pereira et al. 2011), which may enhance the emissions of the two C-based GHGs from BC-added paddy soils upon decomposition. While many studies have shown that biochar addition can suppress CH_4 emissions (Han et al. 2016; Nan et al. 2020; Nguyen et al. 2020), others have reported conflicting findings (Cai et al. 2018; Kimani et al. 2020). Furthermore, BC's effects on CO_2 emissions from submerged paddy soils are limitedly reported. The suppression effects of BC addition on CH_4 emissions may stimulate CO_2 emissions, because the two GHGs can share a similar C source from the GHGs-emitting paddy soils and also because anaerobic oxidation of CH_4 by methanotrophs may lead to increased CO_2 emissions (Kumputa et al. 2019). Nevertheless, limited studies were conducted to assess the trade-off relationship between the two GHGs emitted from the same paddy soils.

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Paddy soils are both an important source and a sink of carbon-based GHGs for the atmosphere (Gupta et al. 2021). A common feature of the paddy soils is predominantly submerged conditions, creating anaerobic conditions by occupying space inside the soil and, therefore, pushing air out of the soil matrices. Under anaerobic conditions, the main electron acceptors for SOC decomposition may include dissolved oxygen (O_2), NO_3^-, Fe (III), and SO_4^{2-} leading to the end product of CH_4 (Sahrawat 2003). On the other hand, CO_2 emissions from anaerobic decomposition of SOC have been reported frequently (Glatzel et al. 2004; Guo et al. 2013; Jiang et al. 2020) and/or from oxidation of CH_4 by methanotrophic bacteria (Kumputa et al. 2019). Consequently, under anaerobic conditions, CH_4 emissions are far greater than CO_2 emissions (Fangueiro et al. 2017; Glatzel et al. 2004). The former can be involved in two microbiological processes of methanogenesis and methanotrophy, both of which can occur strongly under anaerobic conditions (Serrano-Silva et al. 2014). The latter can be linked to both biotic and abiotic processes (Chen et al. 2021; Gougoulias et al. 2014; Hall and Silver 2013; Kumputa et al. 2019; Merino et al. 2021). The biotic processes may require an optimum range of environmental conditions, such as pH and EC for microbial activities (Oertel et al. 2016). The change of these environmental factors may lead to a great reduction of microbial activities and subsequent C-based GHG emissions.

To improve paddy-soil productivity, biochar was introduced and studied. The material has high alkalinity and mineral elements (Fidel et al. 2017; Hossain et al. 2020), which can increase the pH and EC of BC-added soils (Akther 2021; Albert et al. 2021; Hossain et al. 2020). Consequently, BC addition may restrict the activities of CH_4-related bacteria, such as methanogens and methanotrophs, subsequently reducing CH_4 emissions from the BC-added soils (Gupta et al. 2021; Kumputa et al. 2019). The reduction of CH_4 emissions may increase CO_2 emissions, because the two GHGs share a similar C source from SOC. In contrast, the change in environmental conditions caused by BC addition may also lower CO_2 emissions, because emissions are controlled by biotic processes in addition to abiotic processes (Chen et al. 2021; Gougoulias et al. 2014; Hall and Silver 2013; Kumputa et al. 2019; Merino et al. 2021; Zhou et al. 2017). As a result, the effects of biochar addition on CO_2 emissions from paddy soils were reported with inconsistent findings. For example, Qi et al. (2018) found that biochar addition increased CO_2 emissions from paddy soil, compared to the control of no BC addition, while Fidel et al. (2019) reported neutral effects of biochar on CO_2 emissions and Wu et al. (2018) found both neutral and decreasing effects, depending on soil properties. As a consequence, additional studies are needed to determine the co-effects and related mechanisms of BC addition on CH_4 and CO_2 emissions from submerged paddy soils. In addition, the emissions of C-based GHGs can be also dependent on SOC content. Soil with high OC content may release a greater quantity of GHGs than soil with low OC content. The presence of biochar-C may interact with soils of differing OC contents to alter GHG emissions, which needs more studies.

Therefore, the current study, using two paddy soils with different organic carbon contents and biochar produced from corn stalks, was carried out to test the hypothesis that biochar addition can suppress CH_4 emissions while stimulating CO_2 emissions from submerged paddy soils. The aims of the current study were to (1) compare CH_4 and CO_2 emissions and (2) identify some important mechanisms caused by biochar addition in controlling their emissions from two submerged paddy soils.

Materials and methods

Experimental materials

Two paddy soils were taken from two paddy fields in Phuoc Thạnh commune, Cu Chi District, Ho Chi Minh City, Vietnam, (11° 01′ N and 106° 26′ E). The two paddy fields were approximately 700 m apart, and the soils were classified as a Haplic Acrisol (WRB 2015). Field one was located in a relatively lower area with a relatively high SOC concentration (3.05%) and high clay content (21.1%), whereas field two was located in a relatively higher area with a relatively low SOC concentration (0.54%) and low clay content (7.0%) (hereafter called high-OC soil and low-OC soil, respectively). These two fields are located within the rice-growing region of the Phuoc Thanh commune. The climate regime of the examined paddy fields was distinguished by two distinct seasons, the rainy season from May to October and the dry season from December to April. The region had an annual rainfall of around 1868 mm and an average temperature of around 27.4 °C. For soil collection, ten sites were randomly selected from each of the two paddy fields to take soil material from the 0–15 cm layer and the obtained material was mixed well to make a composite sample for each field. Soil sampling from the two selected fields was implemented in 1 day at the end of the rainy season (end of October). The day of sampling was dry but it had rained a few days before. The collected soil was transferred to a laboratory, air-dried, ground, and sieved to pass through a 2-mm sieve to remove plant residues and gravel. Four sub-samples of each composite soil were taken for chemical and physical analysis, and the remainder was used for the incubation experiment.

The biochar used for the current study was produced from corn stalks (*Zea mays* subsp. *mays*), using the method by Nguyen et al. (2018) with a pyrolysis temperature of around 350 °C for about 30 min. The corn stalks were collected from a corn field, chopped into 5-cm long segments, washed

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with tap water, air-dried for a few days, and oven-dried at 70 °C overnight before pyrolysis. The produced biochar was slightly crushed to pass through a 2-mm sieve before the experiment. Some main properties of the biochar and examined soils are shown in Table 1.

Experimental setup

The incubation experiment was set up as a completely randomized design with seven treatments and four replicates. Treatment 1 (T1) consisted of the high-OC soil added with no biochar; treatment 2 (T2) consisted of the high-OC soil added with 2% biochar; treatment 3 (T3) consisted of the high-OC soil added with 4% biochar; treatment 4 (T4) consisted of the low-OC soil added with no biochar; treatment 5 (T5) consisted of the low-OC soil added with 2% biochar; treatment 6 (T6) consisted of the low-OC soil added with 4% biochar, and treatment 7 (T7) was pure biochar (no soil used). T7 was included to serve as a control for comparison to examine the impacts of biochar. These seven treatments were created by combining the ground soils with biochar accordingly. Around 150 g of soil alone (T1 and T4), soil–biochar combination (T2, T3, T5, and T6), and around 20 g of pure biochar (T7) were weighed into 1-L plastic jars with sealing lids fit with rubber septa for gas collection by a syringe. In addition, 0.052-g nitrogen (N) fertilizer, 0.019-g phosphorous (P) fertilizer, and 0.019-g potassium (K) fertilizer (equivalent to 80 kg N, 30 kg P, and 30 kg K applied to the 1-ha paddy field, respectively) were added to each of these jars as applied in paddy rice cultivation. These materials in each jar were mixed well and a plastic net was placed on the surface of these experimental materials in each jar to prevent the biochar from re-floating. Furthermore, a 20-mL glass vial containing 12 mL of 0.5 M NaOH solution was placed in each jar to capture CO_2 emissions. Four 1-L plastic jars containing the four 20-mL glass vials but no experimental materials were also set up to provide background CO_2 and CH_4. All 32 plastic jars (7 treatments × 4 replicates + 4 background jars) were fully immersed in distilled water to a depth of 3 cm. The jars were then tightly sealed and randomly arranged in a laboratory at about 30 °C to start the incubation experiment.

Measurements

Before the experiment, the ground soils and biochar were chemically analyzed for pH, EC, total C, total N, cation exchange capacity (CEC), available P, exchangeable Na, K, and Fe. The materials were added with distilled water at a 1:5 (w/w) ratio, and the extract was measured for pH and EC using a pH meter and EC meter, respectively. The concentration of total C and total N was determined using the dry combustion method with an elemental analyzer (Elementar Analysensysteme GmbH, Hanau, Germany). The CEC was determined using the ammonium acetate method (Carter and Gregorich 2008). The concentration of available P was determined using the Mehlich-1 method ($HCl + H_2SO_4$ extraction) (Novak et al. 2018). The concentrations of exchangeable Na, K, and Fe were measured using the barium chloride method (Carter and Gregorich 2008), and the extract was quantified using inductively coupled plasma–optical emission spectrometry (ICP–OES). In addition, the particle size distribution of the soil was determined (Carter and Gregorich 2008), and the ash content of the biochar was measured by dry combustion at 550 °C overnight. All measurements were done in four replicates, and the mean, as well as the standard deviation of the mean, were calculated and are shown in Table 1.

Air from the headspace of each incubation jar was taken on days 1, 4, 8, 13, 19, 26, 34, 43, and 53 for CH_4 determination. A syringe was used to collect about 15 mL of gas into a 12-mL pre-vacuumed glass vial from 8 to 11 am. The samples were transferred to a laboratory on the same sampling day and analyzed for CH_4 content using gas

Table 1 Selected properties of the experimental materials

Material	Clay %	Silt %	Sand %	pH	EC dS m^{-1}	C %	N %	CEC cmol (c) kg^{-1}	Available P mg kg^{-1}	Na mg kg^{-1}	K mg kg^{-1}	Fe mg kg^{-1}
Mean												
High-OC soil	21.4	19.3	59.3	5.74	0.11	3.05	0.22	11.9	118.5	77.4	147.2	27.6
Low-OC soil	7.0	6.2	86.8	6.13	0.10	0.54	0.05	6.7	156.8	55.1	113.7	17.3
Biochar				9.11	5.15	52.3	0.59	36.9	3046.4	3763	80,752	95.0
The standard deviation of the mean												
High-OC soil	0.27	0.20	0.42	0.06	1.08	0.026	0.005	0.24	1.8	2.5	3.3	0.76
Low-OC soil	0.13	0.17	0.10	0.07	1.83	0.003	0.001	0.62	2.0	2.4	4.4	0.78
Biochar				0.08	0.03	1.005	0.051	0.82	83.4	330	1900	22.6

Ash content of the biochar was 21.5%

CEC cation exchange capacity, $n = 4$

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chromatography–flame ionization detector(Qi et al. 2018). After gas sampling, the lid of each jar was opened, and the 20-mL glass vial was taken out for CO_2 determination by titration method using a 0.25-M HCl solution after precipitating carbonates by about 5 mL of 0.5 M $BaCl_2$ (Zhang et al. 2021). On the same day of gas sampling, the standing water in each jar was also measured for pH and EC using a pH meter and an EC meter, respectively. The pH and EC of the standing water were measured, because the measurements were non-destructive, leading to no significant impact on the ongoing incubation experiment. All jars were opened for around 15 min for air exchange, pH and EC measurements, replacement of the used 0.5-M NaOH solution, and water level checks before re-sealing the lid to start the next incubation period.

Calculation and statistical analyses

The biochar-affected emission ratio (BAER) was computed by dividing the theoretical emission rate (Eq. 1) by the observed amount of corresponding gas. The following equation (Eq. 1) was used to calculate the theoretical emission rate:

$$\mathrm{BAER} = \frac{f_{bc} \times e_{bc} + \left(100 - f_{bc}\right) \times e_{soil}}{100} \quad (1)$$

where f_{bc} is the fraction of biochar added; e_{bc} is the carbon emission rate of pure biochar (treatment 7); e_{soil} is the carbon emission rate of pure soil (treatments 1 and 4). The BAER was used to assess the impacts of biochar on GHG emissions. If the BAER is higher than 1 (theoretical emission rate greater than the observed one), biochar may restrict gas emissions. Otherwise, biochar may stimulate GHG emissions.

Linear and/or nonlinear regression analysis was done to examine the relationship between the measured parameters. All data were statistically analyzed using the Analysis of Variance (ANOVA), following a completely randomized design with four replicates, using JMP 16 (SAS Institute Inc, North Carolina, USA). When the ANOVA result indicated a significant effect at $P \leq 0.05$, the Tukey honestly significant difference test was performed to classify treatment means.

Results

Emissions of carbon-based GHGs

A general dynamics pattern of CO_2 emission rate over the experimental period was a significant increase for the first 8 days, followed by a rapid decrease from day 8 to day 26, and then leveled off (Fig. 1a, b). On day 8, the CO_2 emission rate reached peaked, ranging from 14.3 (mg CO_2–C kg^{-1} soil day^{-1}) in no-BC and low-OC soil (T4) to 32.8 (mg CO_2–C kg^{-1} soil day^{-1}) in 4% BC and high-OC soil (T3). The high-OC soil released more CO_2 than the low-OC soil. Treatments of 4% biochar in two soils (T4 and T6) showed the greatest CO_2 emission rates for all measures when compared to the other treatments of the corresponding soil. The pure-BC treatment also released a considerable quantity of CO_2 with an emission pattern similar to the other treatments. The CH_4 emission rate varied greatly, depending on the examined soil, BC rate, and measurement events (Fig. 1c, d). For the first 4 days in the high-OC soil (Fig. 1c) and the first 8 days in the low-OC soil (Fig. 1d), the CH_4 was released at a lowly increasing rate, but the rate rapidly rose afterward. Of the three treatments in the high-OC soil, no-BC treatment emitted CH_4 at the highest rate in the first six measurements but it emitted at the lowest rate in the last 3 measurements. In contrast, the treatment of 4% BC in high-OC soil had the lowest CH_4 mission rate in the first six measurements but the highest rate in the last measurements. For the three treatments in the low-OC soil, the no-BC treatment had the significantly greatest CH_4 emission rate in measurements 3, 4, 5, and 6 while having the lowest emission rate in the remaining measurements. Treatment of 4% BC in low-OC soil (T6) showed a continuously increasing emission rate of CH_4 from the first to the last measurement. The pure-biochar treatment (T7) had a much lower emission rate of CH_4 in all measurements than the other treatments. The total loss of C as CO_2 and CH_4 was subtracted from the soil organic C and the remaining concentration of SOC was shown in Supplementary Fig. 1. In both soils, the SOC concentration decreased slowly for the first 8 days but rapidly afterward.

Of the six soil-included treatments (T1 to T6), the one applied with 4% BC in high-OC soil (T3) had the greatest cumulative emissions of CO_2 (780), followed by T4 (654), T6 (623), T1 (573), T5 (513), and T4 (334 mg CO_2–C kg^{-1} soil) (Fig. 2a). The high-OC soil emitted significantly greater CO_2 than the low-OC soil and BC addition significantly enhanced CO_2 emissions from the two examined soils. Cumulative CH_4 emissions from T1 (3158), T2 (3128), and T3 (2930 mg CH_4–C kg^{-1} soil) in the high-OC soil were not significantly different from each other, but these from T4 (1255), T5 (699), and T6 (327 mg CH_4–C kg^{-1} soil) were significantly different with each other, with T4 being the highest and T6 being the lowest. The pure-BC treatment (T7) showed the lowest cumulative CH_4 emissions (13.6 mg CH_4 kg^{-1} biochar) of the seven treatments but it had a considerable quantity of CO_2 emissions (671 mg CO_2 kg^{-1} soil), similar to T2 but greater than T4, T5, T6, and T1 (Fig. 2a, b).

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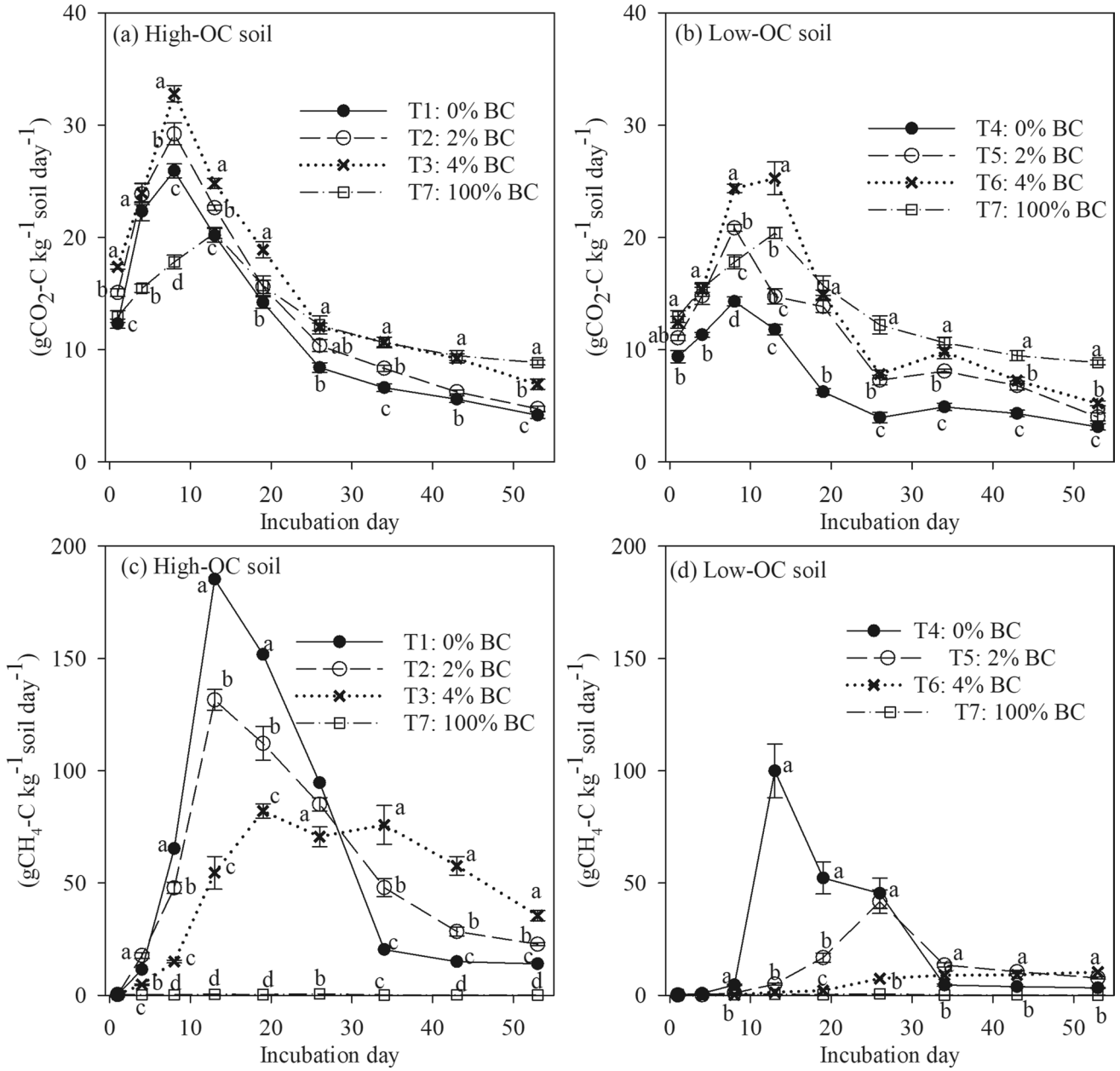

Fig. 1 Dynamics of CO_2 and CH_4 emission rate over the incubation period of 7 experimental treatments. Error bars indicate the standard deviation of the mean. Within a panel and a measurement, bars attached with the same letter are not significantly different from each other. *BC* biochar. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

Relative emissions of GHGs

To investigate the impacts of biochar on emissions of the two GHGs, the biochar-affected emission ratio (BAER, the ratio of theoretical emissions to observed emissions) was computed. The CO_2 BAER of T2, T3, T4, and T6 (Fig. 3a, b) was less than 1 for the whole experimental period, indicating that BC addition increased CO_2 emissions from the BC-added soils. The CH_4 BAER of T2, T3, T5, and T6 (Fig. 4c, d) increased from below 1 in the first few measurements to much greater than 1 in the next measurements, before falling back below 1 at the end of the experiment. T3, T5, and T6 all had BAER values greater than 1 in 5 measurements and T2 had it in 4 measurements.

The GHG emission ratio (GER) of emitted CH_4–C to emitted CO_2–C may suggest a preference for emissions of one GHG over the other. In the high-OC soil, the GER value of high-OC soil added with three levels of biochar (0%, 2%,

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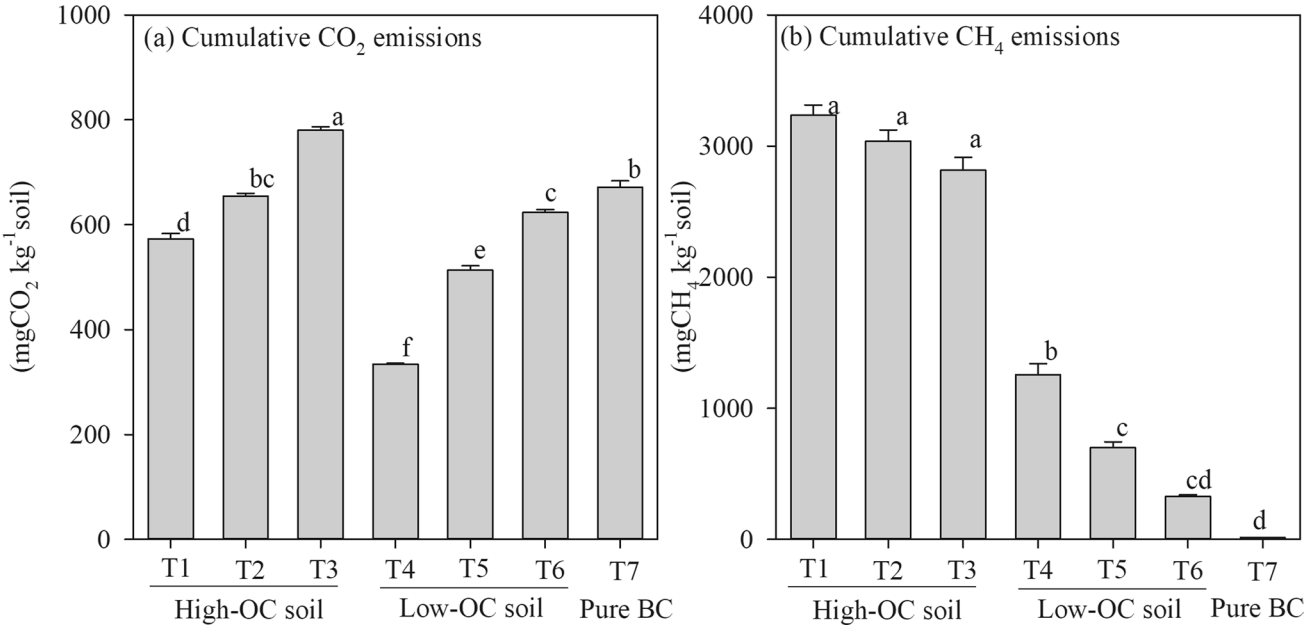

Fig. 2 Cumulative emissions of CH_4 and CO_2 from 7 experimental treatments. Within a panel, bars attached with the same letter are not significantly different from each other. Error bars indicate the standard deviation of the mean. *BC* biochar. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

and 4%) was lower than 1 in the first 2 or 3 measurements, but greater than 1 in the rest measurements (Fig. 4a). In the low-OC soil, 4% BC treatment had a GER of less than 1 in the first 7 measurements, whereas no-BC and 2% BC treatments had GERs of less than 1 in the first 3 and 4 measurements, respectively (Fig. 4b). The remaining measurements were found to have GERs higher than 1. The two GHGs had a negative relationship (Fig. 4c, d). The rise in CH_4 emissions was accompanied by a reduction of CO_2 emissions at rates of -0.26 and -0.29 (mg CO_2–C kg^{-1} soil) for every increasing unit of CH_4 emissions in high-OC soil and low-OC soil, respectively.

pH, EC, and their relationship with GHG emissions

The value of pH and EC of the standing water in each of the 28 incubation jars for all 9 measurements were averaged and shown in Fig. 5. Overall, BC addition resulted in significantly higher pH and EC of the standing water. In the high-OC soil, pH was increased from 6.5 (no biochar) to 7.9 (4% biochar), and EC was raised from 0.49 (no biochar) to 4.7 (dS m^{-1}, 4% biochar). In the low-OC soil, pH increased from 6.8 (no biochar) to 9.3 (4% biochar), and EC was altered from 0.64 (no biochar) to 8.8 (dS m^{-1}, 4% biochar). Pure-BC treatment showed the greatest pH and EC, 10.4 and 15.3 (dS m^{-1}), respectively. The pH and EC values of the low-OC soil were altered more dramatically than these of the high-OC soil.

The pH value of the standing water in the incubation jars was negatively linked with total CH_4 emissions of both soils and was positively correlated with total CO_2 emissions from low-OC soil, but not significantly correlated with CO_2 emissions from high-OC soils (Fig. 6a, b). The correlations between EC value and total CO_2 and CH_4 emissions were comparable to the associations between pH value and the two GHGs (Fig. 6c, d). In high-OC soil, the decreasing pattern of CH_4 emissions over the pH and EC range was well described by a quadratic function, but in low-OC soil, it was well represented by a linear polynomial. A linear function was likewise a good fit for the CO_2 emissions from low-OC soil throughout the EC and pH ranges.

Discussion

An important finding from the current study can be withdrawn from Figs. 1 and 2 that biochar addition suppressed CH_4 emissions while enhancing CO_2 emissions from the two examined soils. Under anaerobic conditions, soil organic matter (SOM) can be decomposed to the final product of CO_2 by heterogenetic respiration using some main inorganic electron acceptors, such as oxidized forms of specific elements Fe, Mn, N, and S (Herndon et al. 2015; Oertel et al. 2016; Sutton-Grier et al. 2011). Consequently, the greater concentration of N and Fe, possibly mainly in the oxidized form due to drying before the experiment, in high-OC soil

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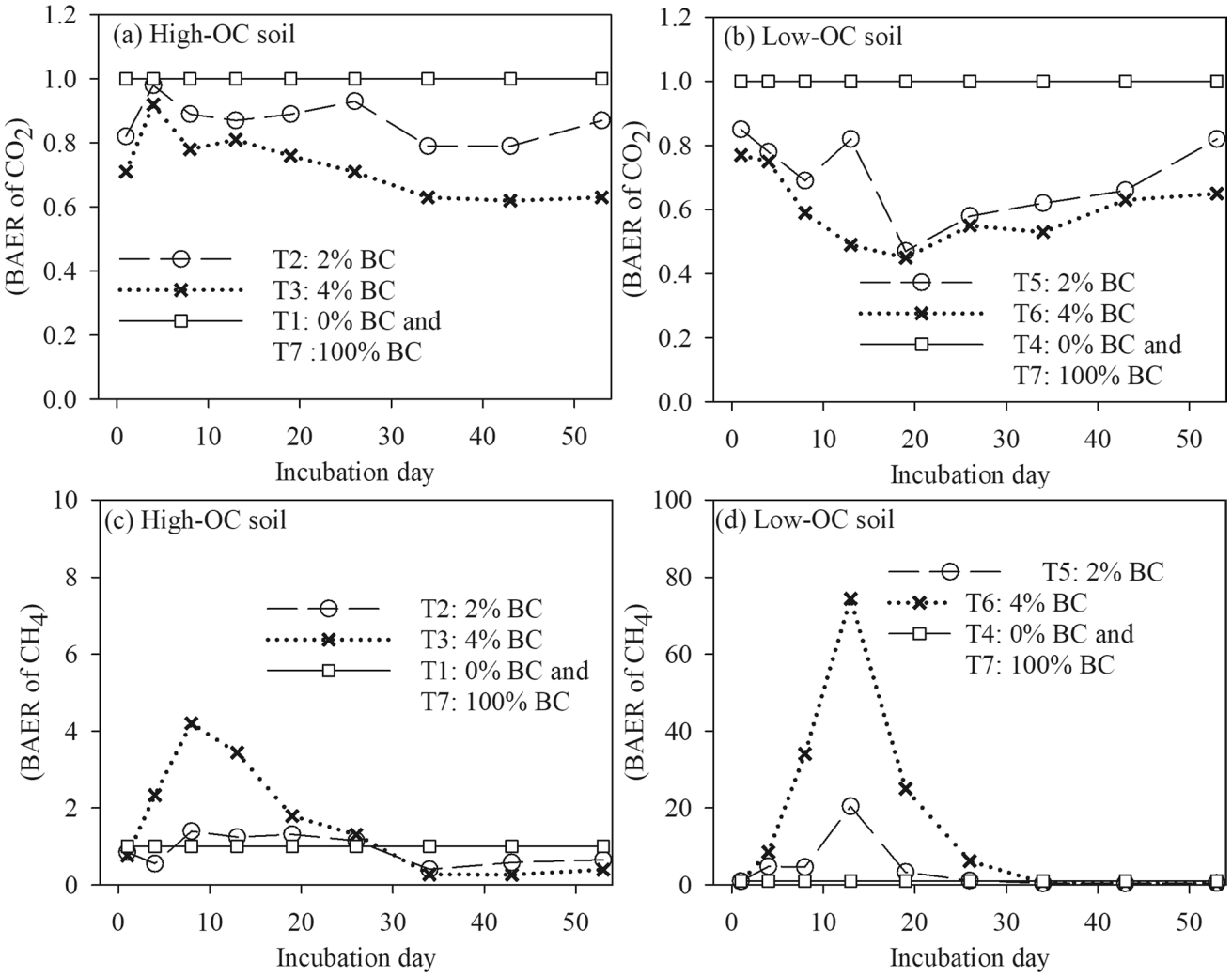

Fig. 3 Biochar-affected emission ratio (BAER) of 7 experimental treatments. Error bars indicate the standard deviation of the mean. *BC* biochar. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

than in low-OC soil (Table 1) could be one cause for greater CO_2 emissions in high-OC soil than in low-OC soil. Furthermore, with a much greater concentration of Fe (Table 1), the added biochar may provide some extra quantity of electron acceptor, such as Fe^{3+} to BC-added soils, promoting SOM decomposition and subsequently CO_2 emissions. The increased CO_2 emissions caused by BC addition could also be because biochar contained a considerable fraction of labile C (Calvelo Pereira et al. 2011; Zimmerman 2010), which was easily decomposed to release CO_2. The reduction of CH_4 emissions by BC addition could additionally enhance CO_2 emissions as the two GHGs share similar C sources from soil (more discussion in following sections).

Commonly, SOM can be oxidized to the final products of CO_2 via mechanisms associated with microbial activities (Gougoulias et al. 2014), which may need an optimal range of environmental conditions to occur. For example, Oertel et al. (2016) reviewed that the pH value for the greatest CH_4 emissions ranged from 4 to 7, while the pH value for CO_2 emissions can be close to the neutral levels. The current study found that BC addition significantly elevated the pH of the examined soils to a level higher than the optimal range. For example, the pH of the high-OC soil increased from 6.5 (no-BC) to 7.9 (4% BC) and that of the low-OC soil was from 6.8 (no-BC) to 9.3 (4% BC) (Fig. 5a). In addition, the EC value of the BC-added soils, which ranged from 2.2 to 8 (dS m^{-1}) (Fig. 5b), might be relatively high, creating an environmental condition unfavorable for microbial activities. Moreover, the pure biochar showed very high pH and EC values of 10.4 and 15.3 (dS m^{-1}), respectively. Nevertheless, CO_2 emissions from soils containing 4% BC and from the pure biochar treatment were significantly greater

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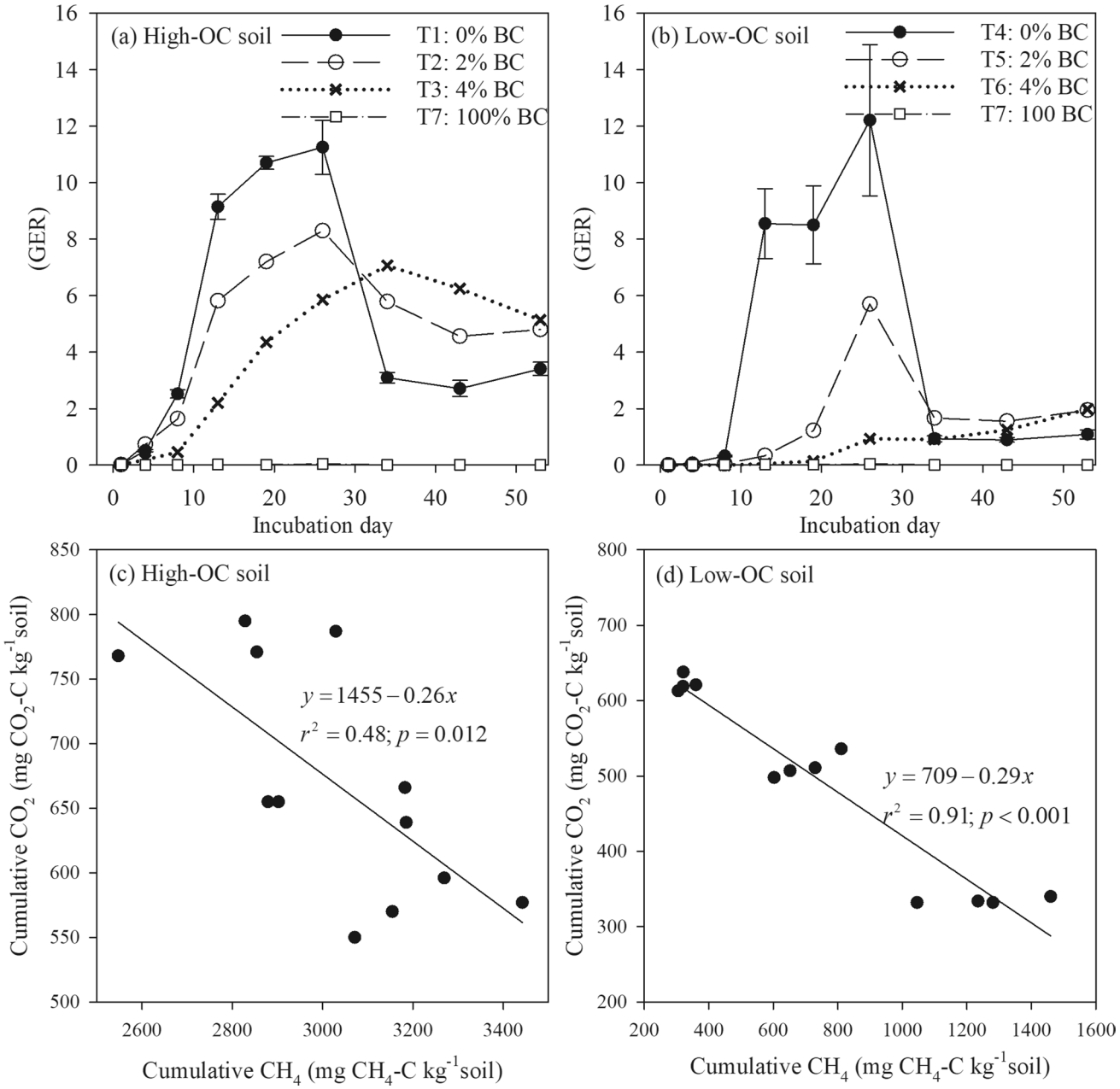

Fig. 4 Gas emission ratio (GER, the ratio of emitted CH_4–C to emitted CO_2–C) and the relationship between the two GHGs of 7 experimental treatments. Error bars indicate the standard deviation of the mean. *BC* biochar. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

than the no-BC treatments (Fig. 2a). These findings may suggest that the mechanisms responsible for CO_2 emissions could not be solely biotic processes but more related to abiotic ones. The abiotic oxidation of SOM was reported by many authors with main mechanisms related to iron oxidation/reduction (Chen et al. 2021; Hall and Silver 2013) and the Fenton reaction (Merino et al. 2021). The hypothesis of the abiotic decomposition of SOM could be additionally strengthened by the impacts of biochar. BC addition may (1) provide BC-added soil with some oxygen contained in the micro-space inside the porous particles (Supplementary Fig. 2) and (2) serve as an electron acceptor for SOC oxidation (Hassanpour et al. 2020) (more discussion on BC effects in the following sections).

In contrast, CH_4 emissions significantly reduced as some environmental properties such as pH and EC of the standing water increased to high levels (10.4 for pH and 15.3 (dS m^{-1}) for EC) by BC addition (Fig. 6). This may suggest

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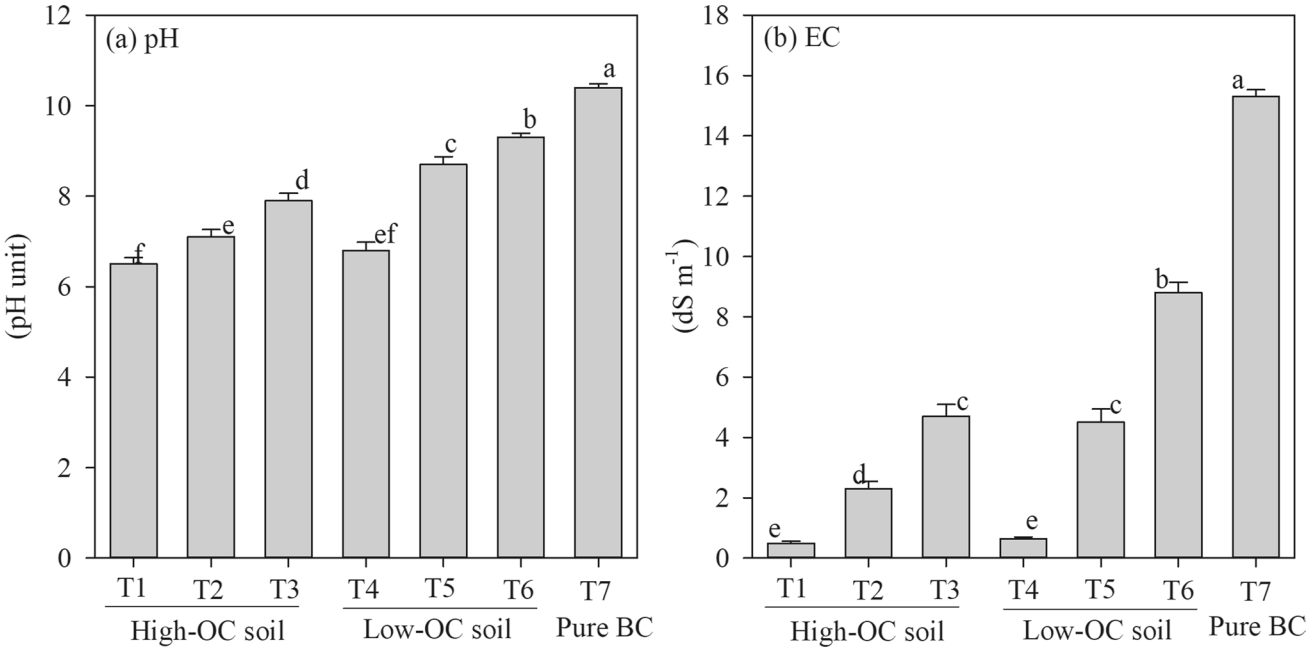

Fig. 5 Average value of pH and EC of 7 treatments. Bars attached with the same letter are not significantly different from the others. Error bars indicate the standard deviation of the mean. *BC* biochar. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

that CH_4 emissions could be more involved in biotic mechanisms induced by two bacterial communities, which are CH_4-generating methanogens and CH_4-consuming methanotrophs (Nan et al. 2021; Sriphirom et al. 2021; Wang et al. 2018). The CH_4-producing bacteria are pH sensitive, and a slight rise in pH over the optimal range (4–7) may suppress CH_4 emission significantly (Oertel et al. 2016; Rahman and Yamamoto 2020). Furthermore, as previously mentioned, the added biochar may introduce more oxygen, toxifying the methane-producing bacteria (Nan et al. 2021). Consequently, biochar strongly restricted the cumulative CH_4 emissions in the current study, as shown in Figs. 2b and 3c, d, by increasing pH and EC values. The negative relationship between CH_4 emissions and EC and pH (Fig. 6) also supported this conclusion. Similarly, biochar addition has been shown to reduce CH_4 emissions from paddy soils (Han et al. 2016; Mohammadi et al. 2020; Nguyen et al. 2020; Yanan et al. 2018) with main biotic mechanisms related to the balance in methanogenic and methanotrophic activities for methane production and consumption, respectively (Han et al. 2016; Nan et al. 2021).

The dynamics of CH_4 emissions can be separated into two stages, the first of which was seen for the first 26 days, and the second was for the last 27 days of the entire experimental period. The first stage was characterized by a greater CH_4 emission rate in two soils added with no biochar (T1 and T4) than in T2, T3, T5, and T6 (BC-added treatments), and the second stage was distinguished by a lower CH_4 emission rate in T1 and T4 than in T2, T3, T5, and T6 for each soil, respectively (Fig. 1c, d). Stronger CH_4 emissions in the first stage and rapidly reduced CH_4 emissions in the succeeding stage in T1 and T4 could be due to the limited labile fraction of SOM, which was quickly depleted after the first stage (Elmira et al. 2013). With a similar explanation based on the hypothesis of the limited labile C fraction, the BC-added treatments (T2, T3, T5, and T6) emitting more CH_4 than no-BC-added treatments (T1 and T4) in the second stage could be due to a relatively higher proportion of the labile C pool remaining after the first stage in the BC-added soil than no-BC soil. This suggested that BC addition prolonged the duration of CH_4 emissions before reaching its maximum rate, as shown in Fig. 1c, d. For example, in high-OC soil, T1 (no BC) and T2 (2% BC) needed 13 days to achieve their CH_4 emission peak; T3 (4% BC) required 19 days; and in low-OC soil, T4 (no BC) needed 13 days; T5 (2% BC) needed 26 days, and T6 (4% BC) needed more than 53 days.

In general, CH_4 emissions need anaerobic conditions to happen, whereas CO_2 emissions may require aerobic conditions or electron acceptors in the anaerobic conditions to occur (Conrad 2020; Deroo et al. 2021). Adding water to the experimental soil may not instantly result in anaerobic conditions, since the experimental materials may contain some oxygen in the micro-space as well as dissolved oxygen in the added water. As a result, CH_4 emissions from the two

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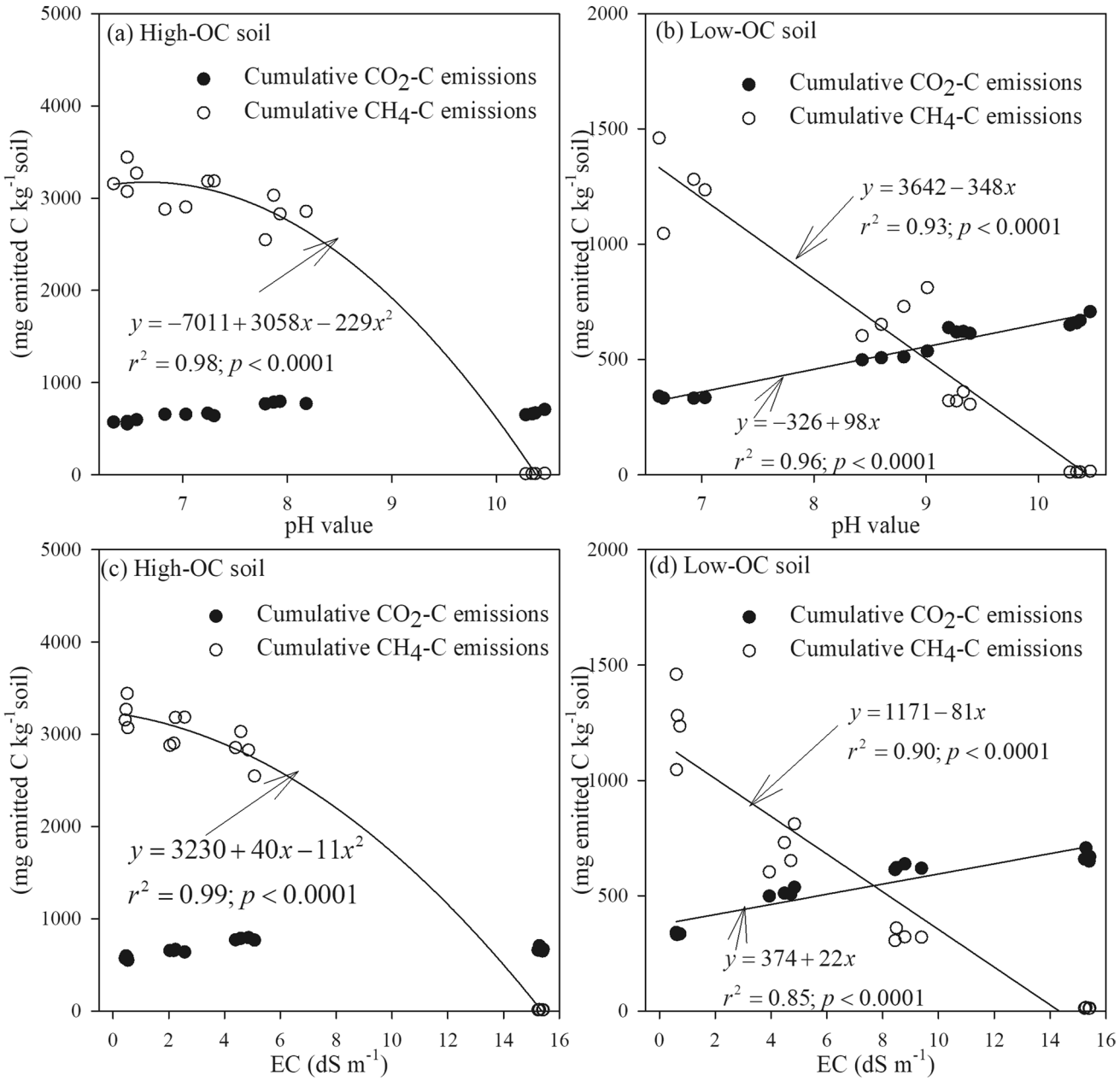

Fig. 6 Relationship between cumulative C emissions and pH and EC of soils added with different biochar rates. T1, T2, and T3 were treatments in high-OC soil added with 0%, 2%, and 4% of biochar, respectively; T4, T5, and T6 were these in low-OC soil added with 0%, 2%, and 4% of biochar, respectively, and T7 was added with 100% biochar

treatments of no-BC addition (T1 and T4) began slowly at a low rate for the first 8 days (Fig. 1c, d), but CO_2 emissions began immediately to rise to their peak after 8 days (Fig. 1). Furthermore, biochar is comprised of porous particles (Supplementary Fig. 2) and may be hydrophobic due to some functional groups, such as the alkyl group (Supplementary Fig. 3) (Mao et al. 2019). Consequently, biochar may still retain a considerable amount of air in the micro-space even after being immersed in water to start the experiment, creating relatively aerobic conditions in some specific micro-sites. Therefore, BC addition may stimulate CO_2 emissions (Fig. 2a) while suppressing CH_4 emissions, in addition to mechanisms discussed earlier.

Because SOC can be the primary C source for released CO_2 and CH_4, soil with a higher organic carbon concentration emitted a greater cumulative quantity of the two C-based GHGs than the soil with a lower organic carbon concentration, as shown in Fig. 2. Emissions of the two GHGs were also dependent on the pH and EC of the standing water (Fig. 6). The value of these two parameters increased

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greatly by biochar addition (Fig. 5). This was likely because biochar itself had high EC and pH values as well as a high concentration of base cations, such as exchangeable Na and K (Table 1). Other studies reported similar findings on increased EC and pH by BC addition (Akther 2021; Albert et al. 2021).

Moreover, biochar had greater impacts on soil properties and GHG emissions in low-OC soil than in high-OC soil. For example, the values of pH and EC of the low-OC soil were elevated to a greater extent than those of high-OC soil (Fig. 5a, b), and the reduced magnitude of CH_4 emissions, as well as the increased extent of CO_2 emissions caused by BC addition, were greater in low-OC soil than in high-OC soil (Fig. 2a, b). These may indicate that high-OC soil could resist the changes caused by BC addition better than low-OC soil. Two factors, that could contribute to the difference in the change-resistant characteristics between the two examined soils, may include SOC content and clay concentration, which were higher in high-OC soil than in low-OC soil. These two factors may lead to a better buffering capacity of high-OC soil than low-OC soil (Curtin and Trolove 2013; Minhal et al. 2019), which may help to mitigate the impacts of biochar in high-OC soil better. In addition, the clay content of high-OC soil (21.7%) was much higher than that of low-OC soil (7.0%, Table 1), possibly leading to greater physicochemical protection of SOC in high-OC soil than in low-OC soil from decomposition (Lützow et al. 2006; Matus 2021).

The CO_2 emissions from the decomposition of SOC under anaerobic conditions were reported by many authors (Glatzel et al. 2004; Guo et al. 2013; Jiang et al. 2020), as well as CH_4 emissions (Nan et al. 2021; Nguyen et al. 2020; Yanan et al. 2018). Because anaerobic conditions may facilitate CH_4 emissions while restricting CO_2 emissions, the current study found that CH_4 emissions from two soils added with no biochar (T1 and T4) were greater than CO_2 emissions (Figs. 1, 2). Nevertheless, the greater CH_4 emission rate than the CO_2 emission rate was also dependent on the experimental treatments as well as the experimental period. For example, in high-OC soil, no-BC and 2% BC treatment needed 8 days to get GERs higher than 1, and 4% BC treatment needed 13 days; in the lower OC soil, no-BC treatment needed 13 days; 2% BC treatment needed 19 days, and 4% BC treatment needed 43 days, while pure biochar treatment had the GER lower than 1 for the whole experimental period. These findings may additionally demonstrate that biochar addition suppressed CH_4 emissions while enhancing CO_2 emissions with many related reasons discussed previously.

For a short-term incubation experiment, the major C source for the two C-based GHGs could be derived from the decomposed labile C portion of SOC. The reduced quantity of cumulative CH_4 emissions caused by BC addition, as shown in Fig. 2, may preserve a considerable portion of the easily decomposable C fraction for CO_2 emissions. This may additionally explain why BC addition significantly increased CO_2 emissions (Fig. 2a) and why CO_2 emissions were inversely correlated with the CH_4 emissions (Fig. 6). Nevertheless, the increased magnitude of CO_2 emissions was less than the reduced magnitude of CH_4 emissions induced by BC addition (Fig. 2), suggesting that the preserved portion of easily decomposable C fraction by BC addition was not entirely decomposed to CO_2 during the experimental time frame. This might imply that, although stimulating CO_2 emissions, BC addition reduced overall C emissions as CO_2 and CH_4 from the submerged paddy soil. Moreover, because CH_4 has a significantly greater global warming potential than CO_2 (Hao et al. 2021), the carbon-negative impact of BC addition on paddy soil could be further increased greatly.

The current study did not quantify the microbial communities involved in the two GHGs. Nevertheless, the current study applied a high BC rate to create environmental conditions unfavorable for the activities of the associated microorganisms. The reduction of CH_4 emission but no response or rising response of CO_2 to the rise in ambient pH and EC as the result of BC addition may strongly suggest that biotic and abiotic processes, respectively, are primarily responsible for the two-gas emissions. The emissions of the two GHGs in the current study were more likely derived from the labile C fraction in the soils. In the long-term experiment or the open-field settings, this C fraction would be quickly deleted. In these situations, the recalcitrant C fraction of SOC may be the main C source for the emissions, which may require more studies. The impacts of biochar on GHG emissions in the open field may be regulated by soil characteristics, such as pH and EC, which are directly influenced by the added BC. As a result, the effects of biochar on C-based GHG emissions could be partly dependent on how soils revert these altered characteristics to their original levels, which may necessitate additional studies.

Conclusions

The emissions of two C-based GHGs from submerged paddy soils were greatly influenced by BC addition and soil properties. In a short-term period, BC addition suppressed CH_4 emissions by −6 to −13% and −44 to −74% while increasing CO_2 emissions by 14–36% and 53–86% in the high-OC soil and low-OC soil, respectively. These two-GHGs emissions were inversely correlated with each other, suggesting that these two GHGs may share a common source of labile C fraction of SOC. The reduced magnitude of CH_4 emissions was much greater than the increased magnitude of CO_2 emissions, indicating that BC addition can have a C-negative impact in submerged paddy soils. BC also substantially raised pH and EC of the examined soils, which may create

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ambient conditions unfavorable for microbial activities. The CH_4 emissions were inversely linked to the pH and EC of the two examined soils, but CO_2 emissions from the low-OC soil were positively correlated with these two parameters, and those from high-OC soil were not connected to either of them. These findings may imply that mechanisms responsible for CO_2 emissions are more likely abiotic, but these for CH_4 emissions can be biotic. In brief, during a short-term experiment in submerged paddy soils, CH_4 emissions were declined but CO_2 emissions were increased, which are more likely associated with the change in pH and EC caused directly by biochar addition.

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Availability of data and materials Data available on request from the authors.

Declarations

Conflict of interest The authors have no conflict of interest to declare.

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