REPORT

Mitigating Global Warming Potentials of Methane and Nitrous Oxide Gases from Rice Paddies under different irrigation regimes

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Abstract A field experiment was conducted in Bangladesh Agricultural University Farm to investigate the mitigating effects of soil amendments such as calcium carbide, calcium silicate, phosphogypsum, and biochar with urea fertilizer on global warming potentials (GWPs) of methane (CH_4) and nitrous oxide (N_2O) gases during rice cultivation under continuous and intermittent irrigations. Among the amendments phosphogypsum and silicate fertilizer, being potential source of electron acceptors, decreased maximum level of seasonal CH_4 flux by 25–27 % and 32–38 % in continuous and intermittent irrigations, respectively. Biochar and calcium carbide amendments, acting as nitrification inhibitors, decreased N_2O emissions by 36–40 % and 26–30 % under continuous and intermittent irrigations, respectively. The total GWP of CH_4 and N_2O gases were decreased by 7–27 % and 6–34 % with calcium carbide, phosphogypsum, and silicate fertilizer amendments under continuous and intermittent irrigations, respectively. However, biochar amendments increased overall GWP of CH_4 and N_2O gases.

Keywords Global warming potential · CH_4 · N_2O · Electron acceptors · Nitrification inhibitor · Rice paddy

INTRODUCTION

Greenhouse gas emissions from rice paddies have been accelerating the global warming, which is of great environmental concern. Among the greenhouse gases, methane (CH_4) and nitrous oxide (N_2O) are the most important due to their radiative effects as well as global warming potentials (GWPs) (IPCC 1995). CH_4 and N_2O gases are simultaneously emitted from rice fields due to their favorable production, consumption, and transport systems between the rice rhizospheric soil and the atmosphere. CH_4 accounts for

15–20 % of the radiative forcing with a GWP of 25 times greater than carbon dioxide (CO_2) on a mass basis (IPCC 2001), while N_2O has a GWP 310 times more than CO_2 and accounts for 6–8 % of the current global warming (Amon et al. 2002). N_2O is naturally produced in soils through nitrification and denitrification (Davidson et al. 1986), which are significantly influenced by type of nitrogenous fertilizer, climate, and soil ecosystem (Akiyama et al. 2006; Synder et al. 2009). It has been reported that N_2O is responsible for the destruction of stratospheric ozone layer (Graedel et al. 1992; Conrad 1996). In irrigated rice, gaseous losses of N may account for up to 48 % of the N applied (Reddy et al. 1980). Bangladesh is a low-lying deltaic country in South Asia, which has a subtropical monsoon climate, e.g., temperature 27–35 °C and high relative humidity 80–90 %. Rice is grown almost throughout the year, which may degrade the environment due to continuous emissions of CH_4 and N_2O gases from rice field. Total rice production in Bangladesh was 34.3 million tons in FY2008–09, where Boro rice contributed about 57 % (18.5 million tons), Transplanted Aman rice 33 %, and Aus rice contributed 10 % (Bangladesh Bureau of Statistics, 2009). Bangladesh will require more than 55.0 million tons of rice to meet the food demand of the expected total population (233.0 millions) by the year 2050. Therefore, rice cultivable area especially boro rice field must be expanded to fulfill the desired production target, which may significantly accelerate CH_4 and N_2O nitrous oxide gas emissions. Rice productivity and carbon storage in paddy soils mainly depend on field management practices such as tillage operations, crop residue management, soil amendments with desirable nutrients fertilizer applications, irrigation water, and drainage systems followed during rice cultivation, which ultimately will affect the rate of CO_2 , CH_4 , and N_2O gas emissions from rice field (Doran and Smith 1987; Balesdent et al. 2000). Among the above options

soil amendments such as silicate fertilization and phosphogypsum application along with nitrogenous fertilizer in rice farming significantly decreased seasonal CH_4 flux by 16–20 % and increased rice productivity by 13–18 % in Korean paddy soil (Ali et al. 2008), whereas 12–21 % reduction in total seasonal CH_4 flux and 5–18 % increase in rice grain yield was recorded through silicate fertilization with urea application in the upland rice paddy soils of Bangladesh (Ali et al. 2012). Another feasible soil amendment is biochar, a charcoal containing high levels of concentrated organic carbon, high porosity (Liang et al. 2006), and greater resistance to microbial degradation in soils (Cheng et al. 2008), which could be introduced in rice paddy soil for improving soil fertility and increasing rice productivity. Besides the potential for improving soil fertility and rice productivity biochar may contribute to mitigate greenhouse gas emissions and increase carbon sequestration in soil (Glaser et al. 2002; Lehmann and Rondon 2006; Dominic et al. 2010; Zhang et al. 2010; Bruun et al. 2011). Calcium carbide (CaC_2), a nitrification inhibitor, could be added with the nitrogenous fertilizers in rice farming to minimize N_2O emissions as well as to improve the nitrogen use efficiency (Bronson and Mosier 1991; Yaseen et al. 2006). Calcium carbide acts as a rich source of acetylene (C_2H_2) gas upon its reaction with water. Acetylene is an effective inhibitor of nitrification and denitrification (Aulakh et al. 2001).

Intermittent irrigations in rice field are expected to decrease CH_4 emission; however, there is a risk of increasing N_2O emissions under well-drained conditions. Soil amendments such as silicate fertilizer and phosphogypsum with high content of electron acceptors along with intermittent irrigations may decrease CH_4 emissions, while addition of nitrification inhibitors, e.g., calcium carbide and biochar with nitrogenous fertilizers may decrease N_2O emissions via controlling the nitrification rate. In the present context of Bangladesh, inadequate information is available about the impacts of soil amendments under different irrigation water management practices on GWPs of CH_4 and N_2O gases. Therefore, this research experiment was undertaken to investigate the effects of the selected soil amendments on CH_4 and N_2O emissions from rice paddy field, and specifically to evaluate the feasibility of decreasing the total GWPs of CH_4 and N_2O gases under the continuous and intermittent irrigations.

MATERIALS AND METHODS

Experimental Site, Soil Amendments, and Rice Cultivation

The experiment was carried out in an ideal paddy field, Bangladesh Agricultural University Farm, Mymensingh in

2010. The soil in the experimental site was silt loam, which belongs to Old Brahmaputra flood plains category. The organic matter content of the soil before experimentation was $39.6 \pm 4.8 \text{ g kg}^{-1}$ and other chemical properties were soil pH (1:5 with H_2O) 6.2 ± 0.2 , available P_2O_5 : $68.9 \pm 2.9 \text{ mg kg}^{-1}$, available SiO_2 $82.6 \pm 3.2 \text{ mg kg}^{-1}$. Two irrigation water management systems such as continuous and intermittent irrigation practices were followed in this experiment. The experimental field was divided into three side by side blocks under each water management regime. Each block had five plots. The area of each unit plot was 100 m^2 . Five treatments such as urea alone (220 kg ha^{-1}), urea plus calcium carbide (30 ppm), urea plus silicate fertilizer (500 kg ha^{-1}), urea plus phosphogypsum (500 kg ha^{-1}), and urea plus biochar (1 t ha^{-1}) amendments were selected in this experiment. The experimental field was laid down in a randomized block design with triple replications. In total, there were 30 unit plots in our experiment; i.e., 2 water regimes * 5 treatments * 3 replications.

All the soil amendments except biochar were applied 2 days before rice transplanting in the field. The biochar was applied 1 week before final land preparation. The biochar used ($<10 \text{ mm}$ sized fraction) in this study was bagasse, a byproduct of sugarcane industry (pyrolysis at $400\text{--}500^\circ\text{C}$ for 2 h). For the field study, the biochar mass was ground to pass through a 2-mm sieve, and mixed thoroughly to obtain a powder consistency that would mix more uniformly with the soil. The contents of total organic carbon (TOC) and total nitrogen (TN) in biochar were 59.7 and 0.65 %, respectively, a total ash content of 20.8 %, and a pH (H_2O) of 9.5. With respect to elemental analysis, the biochar contained 1.0 % Ca, 0.6 % Mg, 0.4 % Fe, and 2.6 % K. The contents of electron acceptors were 1.2, 3.0, 5.5, 4.9, and 4.0 % in urea, urea plus calcium carbide, urea plus calcium silicate, urea plus phosphogypsum, and urea plus biochar amendments, respectively (acid ammonium oxalate in darkness, citrate dithionate extraction method, Loeppert and Inskeep 1996; 2 M Na-acetate extraction method, Kumada and Asami 1958; Loeppert and Inskeep 1996). The basal chemical fertilizers were applied just before rice transplanting: 45 kg N ha^{-1} (urea), $90 \text{ kg P}_2\text{O}_5 \text{ ha}^{-1}$ (super phosphate), and $41 \text{ kg K}_2\text{O ha}^{-1}$ (potassium chloride). Second split of urea fertilizer (35 kg N ha^{-1}) was applied at tiller initiation stage (around 3 weeks after rice transplanting) and the third split of fertilizer (30 kg N ha^{-1} , $17 \text{ kg K}_2\text{O ha}^{-1}$) was applied at panicle initiation stage (around 6 weeks of rice transplanting).

Under the continuous irrigation system, water level in the rice field was kept at 5 cm depth. Under the intermittent irrigation system, rice field was irrigated during the final land preparation to rice transplanting time, active tillering

stage, and flowering stage. The field was kept at moist conditions during the rest of the rice cultivation period. The rice cultivar used in this study was BRRI Dhan 29, indica type, semi dwarf herb with 10–14 tillers, duration 125–133 days. Twenty-one-day-old seedlings of rice cultivar BRRI Dhan 29 (indica type) were transplanted into field plots at a spacing $25 \times 25 \text{ cm}^2$ by the first week of January in 2010 and harvested in the middle of May in the same year.

Gas Sampling and Analysis

A static chamber-GC method (Wang and Wang 2003; Zou et al. 2005a) was used to estimate CH_4 and N_2O emissions during rice cultivation. The air gas samples from the transparent glass chamber (diameter 60 cm, and height 110 cm) were collected by using 60-ml gas-tight syringes at 0-, 15-, and 30-min interval after chamber placement over the rice-planted plots. Gas samplings were carried out once on weekly basis during the rice cultivation. Gas samples were collected three times (8.00–12.00–16.00) in a day to get the average CH_4 and N_2O emissions during the cropping season. The surface area of each chamber was 0.25 m^2 ($0.5 \times 0.5 \text{ m}^2$). While gas sampling, the chamber was placed over six hills of rice vegetation. There were four holes at the bottom of each chamber through which water movement was controlled. Soil and air temperature inside the chamber was recorded for each set of emission measurements. Gas samples in the syringes were stored for analysis by GC in the Laboratory within a few hours. The mixing ratios of CH_4 and N_2O were simultaneously analyzed with a modified Gas chromatograph (Agilent 7890) equipped with a flame ionization detector (FID) and an electron capture detector (ECD; Wang and Wang 2003). A purified gas of nitrogen was used as the carrier gas for CH_4 , and a gas mixture of argon and methane (Ar-CH_4) was used as the carrier gas for N_2O . To remove CO_2 and water vapor in the air samples entering the ECD detector, a filter column filled with ascarite was connected at the beginning of the separation column for N_2O (Zheng et al. 2008). Nitrous oxide was separated by two stainless steel columns (column 1 with 1 m length and 2.2 mm i.d., column 2 with 3 m length and 2.2 mm i.d.) that were packed with 80–100 mesh porapak Q, and detected by the ECD. CH_4 was detected by the FID. The oven was operated at 100°C , the ECD at 300°C , and the FID at 200°C . Fluxes were determined from the slope of the mixing ratio change in three samples, taken at 0, 15, and 30 min after chamber closure. Sample sets were rejected unless they yielded a linear regression value of r^2 greater than 0.90. Average fluxes and standard deviations of N_2O were calculated from triplicate plots. Seasonal amounts of CH_4 and N_2O emissions were sequentially accumulated from the emissions

between every two adjacent intervals of the measurements (Zou et al. 2005a, b).

Estimation of CH_4 and N_2O Emissions

CH_4 and N_2O emissions from paddy fields were calculated from the experimental data and estimated by the following equation at each growth stage (Rolston 1986)

$$F = V/A \cdot \Delta C / \Delta t \cdot 273/T,$$

where F is the methane or nitrous oxide emission rate ($\text{mg m}^{-2} \text{ h}^{-1}$), V is the volume of chamber above soil (m^3), A is the cross-section of chamber (m^2), ΔC is the concentration difference between zero time and time t (mg m^{-3}), and Δt is the time duration between two sampling period (h), T (absolute temperature) = $273 + \text{mean temperature in chamber } (^\circ\text{C})$. The total CH_4 or N_2O emission from paddy fields was the summation of methane and nitrous oxide emissions in all growth stages of rice crop.

Estimation GWPs of CH_4 and N_2O

To estimate the GWP, CO_2 is typically taken as the reference gas, and an increase or reduction in emission of CH_4 and N_2O is converted into “ CO_2 -equivalents” by means of their GWPs. Recently, the net GWP has been estimated to complete understanding the agriculture impacts on radiative forcing (Frolking et al. 2004; Robertson and Grace 2004; Mosier et al. 2006). In this study, we used the IPCC factors to calculate the combined GWPs for 100 years ($\text{GWP} = 25 \times \text{CH}_4 + 298 \times \text{N}_2\text{O}$, $\text{kg CO}_2\text{-equivalents ha}^{-1}$) from CH_4 and N_2O under various agricultural practices. In addition, the greenhouse gas intensity (GHGI) was calculated by dividing GWP by grain yield for rice (Bhatia et al. 2005; Mosier et al. 2006).

Investigation of Rice Plant Growth and Yield Characteristics

Rice plant growth parameters such as plant height, tiller number, leaf area, leaf area index, shoot biomass, root volume, and porosity were investigated during the growing period. Yield components such as panicle number per plant, number of grains per panicle, ripened grains, 1000 grain weight, and harvest index were determined at the harvesting stage. Leaf area was measured by Leaf area meter (Li-3100, Li-COR, USA).

Investigation of Soil Properties

Soil redox potential (Eh) and soil pH were measured by Eh meter (PRN-41, DKK-TOA Corporation) and pH meter (Orion 3 star, Thermo electron corporation), respectively,

during rice cultivation. At the harvesting stage, soil bulk density (BD) was analyzed using cores (volume 100 cm^3 , inner diameter 5 cm), filled with fresh moisture soils. The collected soil core samples were oven dried at $105\text{ }^\circ\text{C}$ for 24 h and then measured the weight of dried core samples. Soil porosity was calculated using the BD and particle density (PD, 2.65 Mg m^{-3}) according to the equation: $\text{porosity (\%)} = (1 - \text{BD/PD}) \times 100$. At harvesting stage, the chemical properties of the collected soil samples were analyzed for pH (1:5 with H_2O), organic matter content (Wakley and Black method; Allison 1965), exchangeable Ca^{2+} , Mg^{2+} , and K^+ (1 M NH_4 -acetate pH 7.0, AA, Shimadzu 660), and available silicate content (1 M Na-acetate pH 4.0, UV spectrometer). The available phosphate content was determined using Lancaster method (RDA 1988). Ferrous iron and water-soluble iron concentrations in fresh soil samples were determined by 2 M Na-acetate extraction method (Modified from Kumada and Asami 1958) and distilled water method (Loeppert and Inskeep 1996), respectively. In the dried soil, the active iron and free iron concentrations were determined by modified acid ammonium oxalate in darkness and citrate dithionite bicarbonate dissolution procedures, respectively, (Loeppert and Inskeep 1996). The dissolved iron and manganese concentrations were quantified by Atomic absorption spectrophotometer (AA Shimadzu 660, Kyoto). The soil $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ contents were determined

acidimetrically after extraction with 1 N KCl (Indophenol Blue Method and by Modified Griss-Ilosvay Method; Keeney and Nelson 1982).

Statistical Analysis

Statistical analyses were conducted using SAS software (SAS Institute 1990). Rice growth and yield parameters, soil properties, methane, and nitrous oxide emission data were subjected to the analysis of variance and regression. Fisher's protected least significant difference (LSD) was calculated at the 0.05 probability level for making treatment mean comparisons.

RESULTS

There were contrasting trends of CH_4 and N_2O emission rates under both continuous and intermittent irrigation systems (Fig. 1). Initially, CH_4 emission rate was slow after rice saplings transplanting in the field until 36 DAT, then gradually increased onwards and sharply increased from panicle initiation stage (63 DAT), peaked ($17.5\text{ mg m}^{-2}\text{ h}^{-1}$) at heading stage (91 DAT) and finally dropped at rice harvesting stage. In contrast to CH_4 , N_2O emissions were very low during the rice growing season. N_2O fluxes showed first peak at active tillering stage

Fig. 1 Trends of CH_4 and N_2O fluxes from continuous and intermittent irrigations with soil amendments during rice cultivation (error bars Standard deviation among the mean values)

(35–43 DAT) following second peak ($35 \mu\text{g m}^{-2} \text{h}^{-1}$) at panicle initiation stage (63 DAT), probably due to splits application of urea which accelerated N mineralization rate, and finally increased N_2O emission rate at the pre-harvesting stage (Fig. 1). Soil amendments such as phosphogypsum, silicate fertilizer, calcium carbide, and biochar in combination with urea were found very effective in controlling CH_4 and N_2O emission rates as compared to those of urea-treated rice planted plots (Fig. 1). Among the amendments silicate and phosphogypsum significantly decreased CH_4 emission rate possibly due to their high content of ferric iron oxides and sulfate ions, being acted as electron acceptors, while N_2O fluxes were decreased remarkably with biochar and calcium carbide amendments probably due to their effects as nitrification inhibitors. The contribution of the soil amendments such as silicate and phosphogypsum having electron acceptors were more pronounced in decreasing CH_4 emission rates under the intermittent irrigation system as compared to continuously irrigated rice field. However, N_2O emission rate significantly increased under the intermittent irrigation system probably due to increased N mineralization rate under

moist dry condition. On the other hand, calcium carbide and biochar amendments were found effective in controlling N_2O emission rate (Fig. 1).

The applied soil amendments contributed to improve the soil redox potential status (Eh) of rice paddy soils and soil pH under both irrigation systems (Fig. 2). Soil pH gradually increased with the application of soil amendments in both irrigation systems, probably due to the release of alkaline materials such as Ca, Mg, and K ions in soil. However, soil redox potential (Eh) value decreased sharply after 35 DAT under continuous irrigation system mainly due to the development of anaerobic conditions and rapid decomposition of soil organic matter. The intense reductive condition, e.g., soil Eh value -100 to -150 mV was observed during flowering to heading stage (77–95 DAT), which caused significant amount of CH_4 emissions (Fig. 2). Soil amendments with intermittent irrigations significantly improved the soil redox potential status and soil porosity, which ultimately decreased CH_4 emissions during the entire rice growing period.

The total seasonal CH_4 flux from the control plots were 124.0 and $90.0 \text{ kg CH}_4 \text{ ha}^{-1}$ from continuous and

Fig. 2 Trends of soil pH and soil Eh under continuous and intermittent irrigations with soil amendments during rice cultivation (error bars Standard deviation among the mean values)

intermittent irrigated rice fields, respectively. Soil amendments, except biochar, significantly decreased total seasonal CH_4 emissions under both irrigation systems. Under continuous irrigation, the total seasonal CH_4 emissions were decreased by 5, 25, and 27 % with calcium carbide, calcium silicate, and phosphogypsum amendments, respectively, whereas under intermittent irrigations CH_4 emissions were decreased by 5, 32, and 38 % with calcium carbide, calcium silicate, and phosphogypsum amendments, respectively. However, biochar amendments increased CH_4 emissions by 5 and 6.0 % under continuous and intermittent irrigated conditions, respectively (Table 1).

The total seasonal N_2O flux from the control plot was $0.550 \text{ kg N}_2\text{O ha}^{-1}$, which was decreased by 47, 36, 33, and 40 % with calcium carbide, calcium silicate, phosphogypsum, and biochar amendments, respectively, under continuous irrigated conditions. On the other hand, the seasonal N_2O fluxes under intermittent irrigated conditions were increased by two folds in all treatments as compared to that of continuously irrigated rice fields. The seasonal N_2O flux from the control plot was $0.98 \text{ kg N}_2\text{O ha}^{-1}$, which was decreased by 30, 21, 19, and 26 % with calcium carbide, calcium silicate, phosphogypsum, and biochar amendments, respectively, under intermittent irrigated conditions (Table 1). The interaction of soil amendments and irrigation water regimes significantly ($p < 0.001$) decreased total CH_4 flux and N_2O flux.

The total GWP of CH_4 and N_2O significantly decreased with soil amendments as compared to those of control plots in both continuous and intermittent irrigations. The total GWP from the continuously irrigated control plot was $3.26 \text{ Mg CO}_2 \text{ ha}^{-1}$ which was decreased by 7, 25, and 27 % with calcium carbide, calcium silicate, and phosphogypsum amendments, respectively, biochar increased total GWP by 1.8 %. Conversely, under intermittent irrigation the total GWP from the control plot was $2.48 \text{ Mg CO}_2 \text{ ha}^{-1}$ which was decreased by 6, 30, and 34 % with calcium carbide, calcium silicate, and phosphogypsum amendments, respectively. Biochar increased total GWP by 4.0 % under continuous and intermittent irrigations. In general, intermittent irrigation system in combination with soil amendments significantly reduced the total GWPs (Table 1), even though N_2O showed two-fold higher GWP in all treatments under intermittent irrigation system compared to those of continuous irrigation system. The GHGI also decreased remarkably with calcium silicate, biochar, and phosphogypsum amendments under both irrigation methods (Table 1).

Soil amendments such as calcium silicate, biochar, and phosphogypsum amendments with urea fertilization significantly increased the panicle number per unit land area which contributed to increased rice yield (Table 1). Rice

grain yield was significantly ($p < 0.01$) increased by soil amendments in both irrigation systems. Using quadratic equation model, the maximum increases in grain yields were estimated 4, 20, 18, and 16 % over the control (4290 kg ha^{-1}) with calcium carbide, calcium silicate, phosphogypsum, and biochar amendments, respectively, under continuous irrigated conditions; whereas grain yields were increased by 6, 25, 21, and 18 % over the control (4350 kg ha^{-1}) with calcium carbide, calcium silicate, phosphogypsum, and biochar amendments, respectively, under intermittent irrigated conditions (Table 1). The concentrations of active iron, $\text{NH}_4^+\text{-N}$, NO_3^- , SO_4^{2-} , and ferrous iron oxides in soil significantly increased with the application of soil amendments in both continuous and intermittent irrigated field plots (Table 2), which acted as electron acceptors in soil and eventually decreased CH_4 fluxes. Higher concentrations of $\text{NH}_4^+\text{-N}$ were observed in the CaC_2 and biochar amended soils as compared to other treatments (Table 2), which acted as nitrification inhibitors in soil and eventually decreased N_2O fluxes. In addition, intermittent irrigation system significantly increased the soil porosity, iron oxides, and the content of soil organic matter (Table 2).

CH_4 flux showed a positive correlation with the availability of soil organic carbon, while there were negative correlations with soil porosity, soil pH, soil Eh, active iron, active Mn, NO_3^- , SO_4^{2-} , and ferrous iron oxides in soil (Table 3). On the other hand, N_2O fluxes showed strong positive correlations with soil porosity, soil nitrate (NO_3^-), and soil Eh, while there were negative correlations with soil ammonium (NH_4) content.

DISCUSSION

In our study, the applied of soil amendments significantly decreased the seasonal CH_4 and N_2O emissions during rice cultivation under both continuous and intermittent irrigation practices. The lower CH_4 emission from the amended paddy soil was due to increased aeration and stabilization of soil C, improved soil redox potential status, higher content of active iron oxides, increased sulfate and nitrate ionic compounds, which acted as electron acceptors, and eventually suppressed CH_4 production. On the other hand, lower N_2O evolution may be an effect of slower N cycling due to inhibition of nitrification and higher C/N ratio with application of selected soil amendments in rice paddy soils. Among the amendments calcium silicate and phosphogypsum significantly decreased CH_4 emission rate due to their high content of active iron oxides and SO_4^{2-} ion which acted as electron acceptors (Inubushi et al. 1984; Yagi and Minami 1990).

Table 1 Rice grain yield, seasonal CH₄ and N₂O emissions, and their GWP_s with soil amendments and irrigation regimes

Irrigation system (A)	Soil amendments (B)	No. of panicles (m ⁻²)	Grain yield (kg ha ⁻¹)	CH ₄ emission		N ₂ O emission		Total GWP (Mg CO ₂ ha ⁻¹)	GHGI ^b
				Seasonal flux (kg CH ₄ ha ⁻¹)	GWP ^a (Mg CO ₂ ha ⁻¹)	Seasonal flux (kg N ₂ O ha ⁻¹)	GWP ^a (Mg CO ₂ ha ⁻¹)		
Continuous	Urea alone	329 ± 13	4290 ± 95	124.0 ± 19	3.10 ± 0.04	0.55 ± 0.09	0.163 ± 0.007	3.26 ± 0.37	0.759 ± 0.05
	Urea + calcium carbide	361 ± 17	4450 ± 107	118.0 ± 17	2.93 ± 0.03	0.29 ± 0.05	0.086 ± 0.002	3.01 ± 0.31	0.676 ± 0.03
	Urea + calcium silicate	445 ± 23	5150 ± 123	93.0 ± 13	2.30 ± 0.02	0.35 ± 0.05	0.104 ± 0.005	2.40 ± 0.25	0.466 ± 0.02
	Urea + phosphogypsum	425 ± 15	5070 ± 117	91.0 ± 11	2.26 ± 0.02	0.37 ± 0.06	0.110 ± 0.007	2.37 ± 0.27	0.467 ± 0.02
	Urea + biochar	419 ± 11	4980 ± 103	130.0 ± 21	2.23 ± 0.02	0.33 ± 0.05	0.098 ± 0.003	3.32 ± 0.39	0.667 ± 0.03
Intermittent	Urea alone	357 ± 17	4350 ± 127	90.0 ± 11	2.19 ± 0.02	0.98 ± 0.19	0.292 ± 0.009	2.48 ± 0.33	0.570 ± 0.03
	Urea + calcium carbide	399 ± 19	4580 ± 139	86.0 ± 9	2.13 ± 0.02	0.69 ± 0.15	0.199 ± 0.007	2.32 ± 0.27	0.506 ± 0.03
	Urea + calcium silicate	462 ± 21	5420 ± 128	61.0 ± 7	1.51 ± 0.01	0.77 ± 0.13	0.226 ± 0.007	1.73 ± 0.21	0.319 ± 0.02
	Urea + phosphogypsum	420 ± 13	5275 ± 116	56.0 ± 6	1.39 ± 0.01	0.79 ± 0.15	0.241 ± 0.008	1.63 ± 0.19	0.309 ± 0.02
	Urea + biochar	441 ± 16	5150 ± 77	96.0 ± 13	2.32 ± 0.04	0.73 ± 0.12	0.217 ± 0.007	2.59 ± 0.37	0.503 ± 0.03
ANOVA	A	***	***	***	***	***	***	***	***
	B	***	***	***	***	***	***	***	***
	A × B	*	*	**	**	**	**	**	*

^a GWP of CH₄ and N₂O were calculated by multiplying 25 and 298 times on seasonal CH₄ and N₂O fluxes, respectively; The IPCC GWP factors (mass basis, kg CO₂-equivalent ha⁻¹) for CH₄ and N₂O are 25 and 298 in the time horizon of 100 years, respectively (Forster et al. 2007)

^b GHGI (kg CO₂-equivalent kg⁻¹ grain yield) was calculated by dividing GWP_s of CH₄ and N₂O emissions by rice yield
*, **, and *** means significant at 5, 1, and 0.1 % probability levels, respectively

Table 2 Physico-chemical properties of soil prior to rice harvesting stage

Soil properties	Continuous irrigation			Intermittent irrigation					LSD _{0.05}		
	Urea alone	Urea + calcium carbide	Urea + calcium silicate	Urea + PG	Urea + biochar	Urea alone	Urea + calcium carbide	Urea + calcium silicate		Urea + PG	Urea + biochar
Soil porosity	0.52	0.54	0.56	0.55	0.56	0.53	0.55	0.57	0.56	0.59	0.01
Soil pH (1:5 with H ₂ O)	6.6	6.8	7.1	6.8	6.9	6.5	6.6	6.9	6.7	6.8	0.03
Soil Eh	−95.0	−73.0	−61.0	−65.0	−68.9	−71.0	−60.0	−43.0	−37.0	−40.0	5.9
Organic matter (g kg ^{−1})	24.5	25.9	28.5	26.9	31.6	24.1	25.1	27.9	26.6	29.5	4.5
Available P ₂ O ₅ (mg kg ^{−1})	63.5	71.5	105.0	98.0	97.0	59.5	67.5	73.5	91.5	89.0	12.0
Available SiO ₂ (mg kg ^{−1})	59.0	79.5	117.0	108.0	93.0	57.5	67.0	97.9	93.0	85.0	15.0
Ex. cations (cmol ⁺ kg ^{−1})											
Ca	3.9	4.1	5.6	6.9	6.3	3.6	3.6	5.7	6.3	5.5	0.35
Mg	1.1	1.3	1.6	1.5	1.5	1.1	1.2	1.4	1.30	1.4	0.11
K	0.21	0.29	0.35	0.38	0.39	0.22	0.34	0.41	0.43	0.47	0.03
Active iron (g Fe kg ^{−1}) ^a	6.50	7.80	11.50	10.20	9.50	6.70	9.20	12.50	11.50	10.30	7.16
Ferrous iron (mg Fe ²⁺ kg ^{−1}) ^b	81.5	123.0	149.0	173.0	143.0	73.5	101.0	117.5	159.0	103.0	29.6
NH ₄ ⁺ -N (mg kg ^{−1})	37.5	49.5	39.5	37.9	35.5	30.7	37.9	28.5	31.5	29.8	4.6
NO ₃ [−] -N (mg kg ^{−1})	0.69	0.83	0.95	0.97	0.93	1.13	0.95	1.07	1.29	1.21	0.31
SO ₄ ^{2−} (mg kg ^{−1})	33.5	57.5	95.0	139.5	121.5	39.5	47.0	129.5	148.5	119.0	23.5

PG phosphogypsum

^a Acid ammonium oxalate in darkness (Loeppert and Inskeep 1996)^b 2 M Na-acetate extractable (modified from Kumada and Asami 1958; Loeppert and Inskeep 1996)

Table 3 Correlation of CH₄ and N₂O emissions with selected plant parameters and soil properties

Parameters (n = 15)	Correlation coefficient (r)			
	Continuous irrigation		Intermittent irrigation	
	CH ₄ emission	N ₂ O emission	CH ₄ emission	N ₂ O emission
Panicle number	−0.584*	−0.456	−0.493*	0.427
Grain yield	−0.547*	−0.643**	−0.641**	−0.601**
Organic carbon	0.189	−0.423	0.175	−0.411
Soil porosity	−0.649**	0.645**	−0.689**	0.868***
Soil Ph	0.656**	0.265	0.743**	0.715**
Soil Eh	−0.753***	0.667**	−0.693**	0.647**
Active Fe	−0.688**	–	−0.665**	–
Ferrous Fe	−0.665**	–	−0.643**	–
NH ₄ ⁺	0.687***	−0.564*	0.567*	−0.669***
NO ₃ [−]	−0.647***	0.746***	−0.623**	0.864**
SO ₄ ^{2−}	−0.798***	0.645**	−0.689***	0.686**

*, ** and *** means significant at 5, 1 and 0.1 % probability levels, respectively

On the other hand, N₂O fluxes were decreased remarkably with biochar and calcium carbide amendments due to their effects as nitrification inhibitors. Slow decay of biochar in soils together with tillage and transport activities may release a small amount of CH₄ and CO₂ to the atmosphere. The carbon in biochar prevented soil degradation and hold C in soil for long period of time (Bruno et al. 2002). It has also been shown that biochar-amended soil retains ammonium (NH₄⁺) and nitrate (NO₃[−]) ionic compounds (Liang et al. 2006; Cheng et al. 2008). Johannes (2007) reported that biochar amendments in soil substantially reduced nitrous oxide emission, probably due to the increased synchrony in the mineral N availability and plant uptake. The concentrations of active iron oxides (Fe₂O₃), NH₄⁺, NO₃[−], SO₄^{2−}, and ferrous iron oxides significantly ($p < 0.001$) increased in the amended paddy soil (Table 2). Seasonal CH₄ emissions showed strong negative correlations with the active iron, SO₄^{2−}, and ferrous iron concentrations in soil at harvesting stage (Table 3). This implies that the released iron oxides and sulfate from the applied soil amendments acted as electron acceptors and eventually suppressed CH₄ emissions as supported by Jackel and Schnell (2000). On the other hand,

seasonal N₂O emissions showed strong positive correlations with the soil porosity, NO₃[−], soil pH, and soil Eh (Table 3). Bronson et al. (1991) reported that the slow release of acetylene from urea-encapsulated calcium carbide significantly reduced CH₄ and N₂O emissions during rice cultivation and increased rice yield. Rice grain yield was negatively correlated with seasonal CH₄ flux (Table 3), which was supported by Denier van Der Gon et al. (2002). The increased carbon sources in paddy soil enhanced CH₄ emissions but decreased N₂O emissions were observed under anaerobic conditions (Hou et al. 2000). Zhang et al. (2010) reported that biochar amendments with 10–40 t ha^{−1} increased rice yields by 12–14 % in unfertilized soils and by 9–12 % in soils with N fertilization, respectively. They also found that total soil CH₄–C emissions were increased by 34 and 41 % in soils amended with biochar at 40 t ha^{−1} compared to the treatments without biochar and with or without N fertilization, respectively. However, total N₂O emissions were sharply decreased by 40–51 % and by 21–28 %, respectively, in biochar amended soils with or without N fertilization. Wang et al. (2011) reported that biochar incorporation (50 t ha^{−1}) into paddy soils significantly decreased N₂O emissions during the 60-day period by 73 %, while the inhibition ranged from 51 to 94 % ($p < 0.05$ –0.01) in terms of cumulative emissions. Wang et al. (2012) reported that biochar application decreased N₂O emissions up to 54 and 53 % during rice and wheat seasons. However, Clough et al. (2010) and Kristiina et al. (2011) found that biochar application had no effect on N₂O emissions. Zhang et al. (2012) reported that biochar amendment (20 t ha^{−1}) in a Chinese paddy soil increased GWP by 39 % in the first growing cycle; however, the overall GWP decreased (7–18 %) in the second consecutive growing season.

Soil amendments improved soil redox potential status and soil porosity under intermittent irrigation, provided a soil condition favorable for N₂O production primarily through nitrification followed by denitrification (Hou et al. 2000). Soil pH was shifted toward neutral point (7.0–7.1) in the paddy rice field, which was favorable to the growth of methanogenic bacteria and other microbial organisms. The seasonal N₂O emissions were low in all treatments under continuous irrigation as compared to those of intermittent irrigation, which may be due to less nitrification activities under anaerobic condition. Seasonal N₂O emissions were increased by 16–43 % with intermittent irrigations compared with continuous flooding.

The N₂O emission peaks were observed at 35 and 63 DAT, probably due to the split applications of nitrogenous fertilizers and maximum availability of mineral N until the middle season. Afterward a small flux of N₂O was recorded in the late season. Draining the rice fields around

60–65 DAT increased O₂ availability in the soil for N₂O production as the intermediate product of either nitrification or denitrification (Xiong et al. 2007; Zheng et al. 2008). Moreover, midseason drainage improved root activities and accelerate soil organic C decomposition, which might produce more available C and N for soil microbes, and thereby favor N₂O emissions (Zou et al. 2007). Rondon et al. (2005) observed a 50 % reduction in N₂O emissions from soybean plots and almost complete suppression of CH₄ emissions from biochar-amended (20 Mg ha⁻¹) acidic soils in the Eastern Colombian Plains, possibly due to better aeration and greater stabilization of C. The lower N₂O evolution may also be an effect of slower N cycling possibly due to a higher C/N ratio. Yanai et al. (2007) observed 85 % reduction in N₂O production of rewetted soils containing 10 % biochar compared to soils without biochar. Roberts et al. (2010) reported that 30 % GHG emissions (−864 kg CO₂e Mg⁻¹ dry corn stover) could be reduced with biochar application to soil rather than combusted for energy generation.

In our study, maximum level of NH₄⁺-N (43.7 mg kg⁻¹ soil) was observed in the CaC₂ amended soils before rice harvesting (Table 2), which could be due to inhibition effect of CaC₂ for nitrification and eventually caused minimum N₂O production. Kashif et al. (2007) reported maximum concentration of NH₄⁺-N (41.2 mg kg⁻¹ soil) in the CaC₂-amended (30 mg kg⁻¹ soil) soil receiving 60 mg N kg⁻¹ as urea, while lowest concentration of NH₄⁺-N 14.3 mg kg⁻¹ was recorded in the absence of calcium carbide. The application of encapsulated CaC₂ significantly suppressed the oxidation of NH₄⁺-N to NO₃⁻-N in urea-fertilized soil, being supported by other researchers (Yaseen et al. 2006). Among the amendments silicate fertilizer and phosphogypsum, being industrial byproducts are easily available, economically feasible, and effective for mitigating seasonal CH₄ flux and increasing rice productivity, while calcium carbide and biochar amendments in selected paddy soils seem to be effective for reducing N₂O emission.

CONCLUSION

Calcium carbide, calcium silicate, and phosphogypsum amendments in paddy soil decreased total GWPs of CH₄ and N₂O gases by 6–34 %, whereas biochar amendments increased total GWP by 4.0 %. Rice grain yield was increased by 16–20 % over the control with biochar, phosphogypsum, and silicate fertilizer amendments with conventional urea fertilizer application. Among the amendments, phosphogypsum and silicate fertilizers were found effective for suppression of CH₄ emissions, while calcium carbide and biochar for reducing N₂O emissions

from the flood plain paddy soils in Bangladesh. As the emission of CH₄ and N₂O gases from paddy soils depends on the inherent characteristics of soils, exogenous N-fertilizer sources, microbial activity, and soil ecosystem, therefore, economically viable biochar production technology and regional trials to different agro-ecological zones require further research.

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