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RESEARCH ARTICLE

EFFECT OF FERTILIZER ON METHANE AND NITROUS OXIDE EMISSIONS FROM RICE-CROPPING SYSTEM AT TWO LOCATIONS OF INDO-GANGETIC REGION OF WESTERN UTTAR PRADESH, INDIA

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ABSTRACT

Agriculture soils of any country significantly contribute to the greenhouse effect primarily through the emissions of greenhouse gases (GHG'S) like methane (CH₄), nitrous oxide (N₂O) and carbon dioxide (CO₂). The climate change risk in Asia may impact Indo-Gangetic plains (IGP) which may be affected greatly by the melting of Himalayan glaciers. The dominant land use by farming community in the IGP is rice-wheat cropping system. CH₄ emission depends on crop, growth season, fertilizer application and prevailing environmental conditions of the region. Various agricultural soils, climate and farm management practices control the CH₄ and N₂O emissions. CH₄ and N₂O from rice fields are prominent greenhouse gases which are emitted from paddy fields. The present investigation was carried out through close chamber techniques to find out the variation in CH₄ and N₂O fluxes at two different locations of IGP region during 2012. The value of CH₄ flux (Kg ha⁻¹ d⁻¹) from rice crop varied from 0.348 to 1.343 at Chachula and 3.794 to 13.686 at Meerapur while the flux (g ha⁻¹d⁻¹) varied from 0.30 to 7.9 at Chachula and 0.60 to 8.5 at Meerapur for N₂O respectively. Appropriate application of fertilizer and irrigation in lowland rice field also decrease nitrous oxide emission. It is observed that the emissions of CH₄ as well as that of N₂O are increased with the application of commercial fertilizer in both the locations of Chachula and Meerapur from western Uttar Pradesh of Indo-Gangetic region. Statistical comparisons of CH₄ and N₂O flux were also performed with one-way analysis of variance (ANOVA).

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INTRODUCTION

The concentration of trace gases like CH₄ (1774 ppb) and N₂O (319 ppb) have increased in the atmosphere exponentially as a result of modern human living. The higher concentration of CH₄ and N₂O in the earth atmosphere is predominantly due to agricultural activities and increasingly use of fossil fuel. Globally, agriculture activities contribute to the tune of approximately 60 % of nitrous oxide and 50 % of methane emissions. Agricultural CH₄ and N₂O emissions have increased by 17 % between 1990 to 2005 (Smith *et al.*, 2007). The three major sources of global methane and nitrous oxide emission from the agriculture sector are soil (38% of CH₄+N₂O), paddy production (11% of CH₄) and biomass burning (12 % of CH₄+N₂O). Methane, Carbon dioxide, Nitrous oxide and Chlorofluorocarbons (CFCs) are the keys greenhouse gases, which have strong infrared absorption bands and trap part of

the thermal radiation emitted from the surface earth. The concentrations of carbon dioxide, methane and nitrous oxide in the atmosphere have increased from 337 to 360, 1.50 to 1.72, 0.302 to 0.320 ppmv respectively, during last decade (Rasmussen and Khalil, 1986; Battle *et al.*, 1996). Anaerobic decomposition of organic matter in flooded paddy fields produces methane (CH₄), which escapes to the atmosphere primarily by mechanisms of diffusive transport through the paddy plants during the growing season. Paddy fields, which are not flooded and therefore do not produce significant quantities of CH₄, account for approximately 10 % of the global paddy emission and about 15 % of the global paddy area under cultivation. The remaining area is grown adopting wetland rice, consisting of irrigate and deepwater rice cultivation. The global wetland paddy area harvested annually in the early 2010 was about 123.2 million hectares (Mha) out of 144 Mha harvest area of paddy, over 90 per cent of which was in Asia (Neue *et al.*, 1994). Fourteen of the wide variety of sources of atmospheric CH₄, rice paddy fields is considered one of the most important. The Intergovernmental Panel on Climate Change (IPCC, 1996) suggested the global emission

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rate from paddy fields at 60 Tg/yr, with a range of 20 to 100 Tg/yr. This estimate is about 5-20 per cent of the total emission from all anthropogenic sources. This figure was arrived mainly based on field measurements of CH₄ fluxes from paddy fields in the United States, Spain, Italy, China, India, Australia, Japan and Thailand. The measurements at various locations of the world indicate that there are large temporal variations of CH₄ fluxes and that the flux differs markedly with soil type, texture and with application of organic and commercially mineral fertilizers (Neue and Sass, 1994). The wide variations in CH₄ fluxes also exhibit that the flux is critically dependent upon several factors including climate, characteristics of soils paddy agricultural practices, particularly water regime. The parameters that affect CH₄ emissions from paddy field vary widely both spatially and temporally. Multiple year data sets near the same location and under similar conditions can lead to substantial differences in seasonal methane emission levels, making it difficult to establish a single number as the emission level from a field. Thus, at the current level of knowledge, a reported range in CH₄ emission levels for a country is more realistic than a single number. The major pathways of CH₄ in high wet soils are the reduction of CO₂ with H₂, with fatty acids or alcohols as hydrogen donor, and the transmethylation of acetic acid or methanol by methane-producing bacteria (Takai and Wada, 1990; Conrad, 1989). In paddy fields, the kinetics of the reduction processes is strongly influenced by the composition and texture of soil and its content of inorganic electron acceptors. The period between flooding of the soil and the onset of methanogenesis can apparently differ for the various soils. However, it is not obvious if soil type also influence in the field methanogenesis and CH₄ emission when steady state conditions have been obtained (Conrad, 1989).

The redox potential (Eh) is one of the key parameter for generation of CH₄ in soils. The Eh, or electron activity, of the soil gradually decreases after flooding. Patrick and Reddy (1978) demonstrated that the redox potential of a soil must be below approximately -150 mV in order to have CH₄ production. Yamane and Sato (1964) also showed that the evolution of CH₄ from flooded paddy soils did not commence until the Eh fell below -150 mV. It has been widely observed that the Carbon substrate and nutrient availability are also important factors. That straw to paddy fields significantly increases the CH₄ emission rate compared with application of compost prepared with rice straw or chemical fertilizer. Soil temperature is known to be an important factor in affecting the activity of soil microorganisms. This is to a certain extent related to the soil moisture content because both the heat capacity and the heat conductivity are lower for a dry soil than for a wet soil. Yamane and Sato (1961) have shown that CH₄ formation attained a maximum value at 35°C in waterlogged alluvial soils. The rate of methane formation was significantly small below 20°C. Since the conversion rate of substrate to CH₄ depends on the temperature it is observed that the momentary local emission of CH₄ from the soil to the atmosphere depends on the temperature. However, the dependence of the seasonally integrated emissions of CH₄ on temperature is much weaker. The emission of CH₄ may also depends primarily on the total input of organic substrate: although the temperature determines the time it takes to convert the substrate to CH₄, that time is generally short compared to a season. Therefore, the methodology adopted here is based on the seasonally and integrated CH₄ emission, whose temperature dependence can be neglected in first approximation. It is generally believed that the CH₄ formation

is only efficient in a neutral pH range (pH from 6.4 to 7.8). The effect of flooding in the field is to increase the pH in acid soil on other hand it decreases the pH in alkaline soil. The rise of pH in acidic soils is due to the reduction of acidic Fe³⁺ to Fe²⁺ which simultaneously reduces the presumably potential Eh. The addition of nitrate as chemical fertilizer to flooded soils may suppress the production of CH₄, because nitrate acts, as well as Fe³⁺, Mn⁴⁺, as a terminal electron acceptor in the absence of molecular oxygen during anaerobic condition, and poises the redox potential of soils at values such that the activity of strict anaerobes is restricted. The addition of sulphate may also inhibit CH₄ output for similar reasons as nitrate. There are three ways of CH₄ release into the atmosphere from rice fields. Emission through diffusion loss of CH₄ across the water surface is the least important process. Methane loss as bubbles (ebullition) from rice fields is established and significant mechanism, especially if the soil texture is not clayey. During land preparation and initial rice growth, ebullition is the major release mechanism. The third important process is CH₄ transport through rice plants stem, which has been reported as the most notable phenomenon (Seiler *et al.*, 1984; Schütz *et al.*, 1989b). Several researchers have demonstrated that more than 90% of total CH₄ emitted during the cropping season is released by diffusive transport through the aerenchyma system of the rice plants and not by diffusion or ebullition. Emission through rice plant may be expected to show significant seasonal variations as a function of changes in soil conditions and variations in plant growth.

The CH₄ emission rates are function of the partial pressure of CH₄ in the soil. Part of the CH₄ produced in the soil is taken up by the oxidized rhizosphere of rice roots or by the oxidized soil-floodwater interface. It is considered that the soil methanotrophic bacteria can grow with CH₄ as their sole energy source, and other soil bacteria, such as *Nitrosomonas* species are also able to consume CH₄ (Conrad, 1993). CH₄ is also leached to ground water, as a small part and is dissolved in water. Therefore, reduction in soil CH₄ does not necessarily mean that all CH₄ has been emitted into the atmosphere.

CH₄ Emission Studies in India

The estimates of CH₄ budget from Indian paddy fields are of special significance as India has 42.2 million ha of land under rice cultivation, of which 16.4 million ha is irrigated and the remaining is rained (19.7 and 5.9 million ha lowland and upland, respectively). Several studies have been carried out on CH₄ emission from rice fields in India. The seasonal CH₄ emissions were observed to be 16-630, 4-109, 37-530 and 0.1-1650 kg/ha in eastern, central, southern, and northern India, respectively. Wide variations in CH₄ emission are due to variations in soil organic carbon, texture, pH, and other physico-chemical properties and different agronomic practices, including fertilizer and water management and cultivar of rice used at these locations. In 1991, the United State Environment Protection Agency (USEPA) reported that 37.8 Tg/yr of CH₄ was emitted from rice growing regions of India. A broad measurement campaign (1989-1991) covering selected rice growing regions of India however, indicated a very low source strength ranging from 3.64±1.26 Tg/yr. According to (Watson *et al.*, 1992), CH₄ emission from rice paddies in India is 2.4-6 Tg/yr. However, more recently Pathak *et al.*, 2003 have estimated that total methane emission from Indian rice fields is 2.9 Tg/yr. In India, range of CH₄ flux values varied between 0.20-3.6, 0.04-6.6 and 1.1-23.3 mg/m²/d⁻¹ for irrigated and

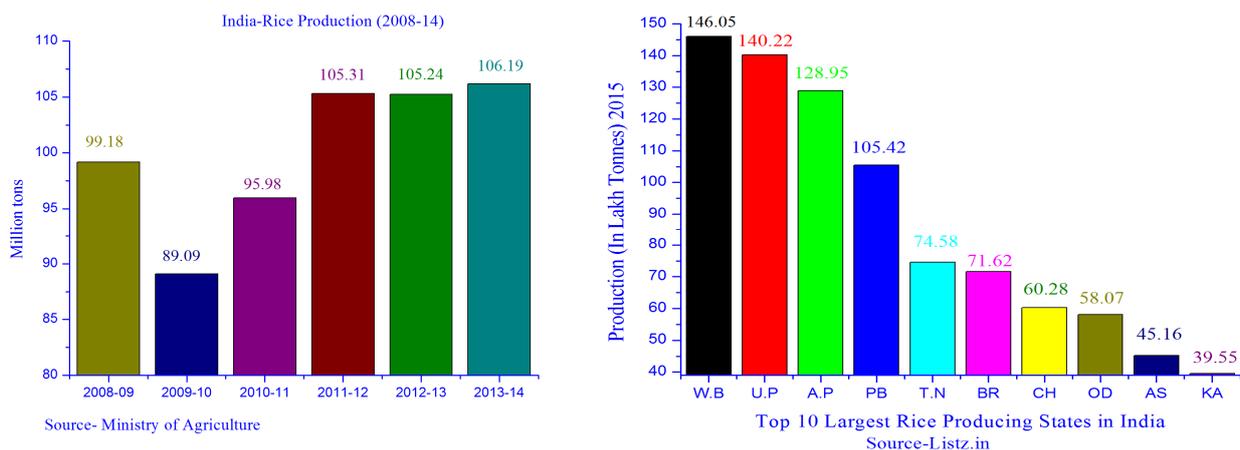


Fig.1. Rice production from 2008-2014 with top ten rice producing states in India (2015)

intermittently flooded, flooded and deep-water rice fields, respectively. These estimates, however, require further refinements because of uncertainties caused by scarcity of flux measurements data from larger areas, gaps in the knowledge of rice ecologies, the impact of soil types, crop management and lack of data on in situ CH_4 oxidation.

Estimates of CH_4 Emission

The estimates on CH_4 emission from paddy fields have varied considerably over the time horizon. Enhalt and Heidt, 1973 estimated it to be 280 Tg/yr. Cicerone and Shetter, 1981 gave the revised estimates for CH_4 emission at 59 Tg/yr. Another estimate of the global emission of methane from rice fields is in the range of 30-70 Tg/yr based on various model calculations by different groups. The recent estimates of the International Panel for Climate Changes (IPCC) using Special Report on Emissions Scenarios (SRES) are around 300 Tg in 2000, and between 400 and 600 Tg in 2010. Measurements in rice paddies in various locations of Asia show that there are large temporal variations of CH_4 emissions differing markedly with climate, soil and paddy characteristics, fertilizers applied, organic matter and other agricultural practices. These observations indicate the average emission range of methane flux from 18.4-1540 kg/ha/yr. Further CH_4 emission was reported as low as 4 kg/ha/yr in IRRI, Philippines and the highest being 2110 kg/ha/yr in Shenyang, China. Neue and Scharpenseel, 1984 reported that irrigated, rainfed and deep-water rice contributes 75, 22, and 3 percent of the total global CH_4 emission from rice fields, respectively. Several reported studies also show complex nature of variations in seasonal methane emission from rice paddy fields. Also some studies have reported a correlation with soil temperature (Schütz *et al.*, 1989a,b), but not reported in others (Cicerone *et al.* 1983; Neue and Sass, 1994). Most of the CH_4 emitted from rice fields is expected to be from Asia, as it has 90 % of the total world rice harvested area, out of which about 52 percent falls in China and India.

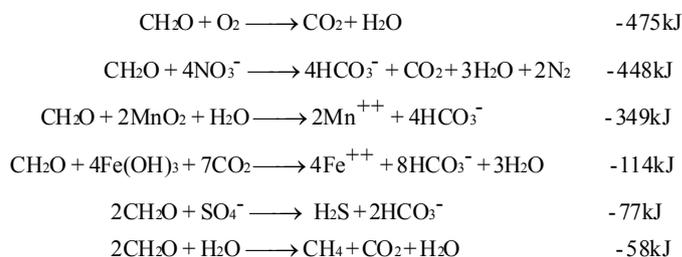
Rice Fields as a Source of CH_4

Isermann, K. 1994 carried out first *in situ* measurement of the CH_4 flux in California, USA by Cicerone and Shetter, 1981 followed by extensive studies in other parts of the world. The field experiments stressed the importance of rice plant as a conduit pipe for CH_4 transport from soil to the atmosphere. At present the CH_4 source strength of wetland rice fields is

estimated at 60 Tg per year, with a range of 20-100 Tg per year. However, this estimate is still tentative and efforts are being made to make it more realistic. International Panel on Climate Change (IPCC) has started a worldwide campaign to update the inventory of CH_4 emission from various sources.

Methanogenesis

Methanogenesis, the biological production of CH_4 , is a geochemically significant process that occurs in all-anaerobic environments in which organic matter undergoes decomposition. The biogenic CH_4 results from the metabolic activities of a small and highly specific bacterial group, which are terminal members of the food chain in their ecosystem and are called methanogens. Methane is produced by the reduction of soil organic carbon by methanogens process under strict anaerobic conditions having redox potential of less than -150 mV. When soil is under oxidized environment, aerobic decomposition occurs with the consequent release of carbon dioxide. Anaerobic conditions appear in wetland rice fields as a result of soil submergence. Water saturation of soil limits the transport of O_2 into the soil. Under such anaerobic condition, microorganisms start using alternative electron acceptors in their respiration causing further soil reduction. The redox potential drops sharply in a sequence, eventually leading to methanogenesis. The sequence of the redox reactions is as follows:



The reaction sequence is strictly in accordance with the yield of free energy. The process of soil reduction tends to stabilize the soil pH near neutral, which is optimal for methanogenesis. High salinity and sulphate concentration increase competitive interactions of sulphur reducing bacteria and methanogens. Application of organic manure and fertilizers and submergence of soil with deep water increases the population and activities of methanogenesis bacteria in the paddy fields. With the onset of anaerobic and reduced conditions, methanogens produce CH_4

from either the reduction of CO₂ with H₂ (hydrogen trophic) or from the fermentation of acetate to CH₄ and CO₂ (acetoclastic). In nature the later mechanism accounts for about two-third of the CH₄ emission from soil. Under steady-state conditions in anoxic rice fields the acetoclastic pathway is dominant and accounts for about 75-80 percent of the total CH₄ emitted. These bacteria being strictly anaerobic, convert fermentation products formed by other microorganisms, notably CO₂, H₂, esters, and salts of formic acid (HCOOH) into CH₄, but other substrates may be used as well. The reactions with reference to the type of methanogens involved in forming CH₄ as an end product can be illustrated as.

a) H₂ reduction of CO₂ by chemoautotrophic methanogens:



b) Several strains of methanogens can also use HCOOH or CO as a substrate for producing CH₄ in addition to CO₂ and H₂:

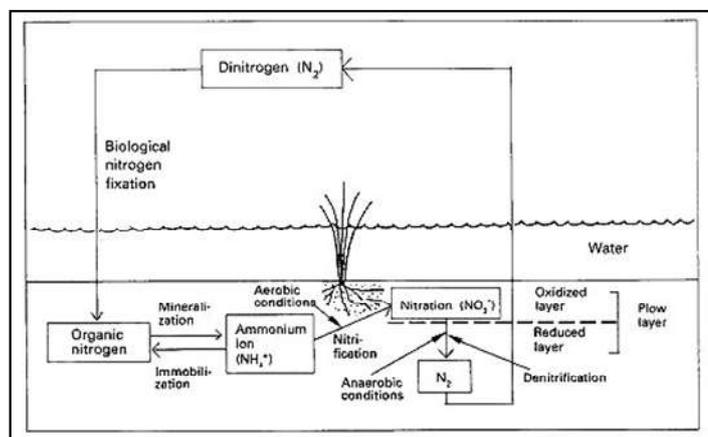


c) Methane can also be produced by methyl trophic methanogens, which use methyl-group containing substrates such as methanol, acetate and trimethylamine:



Rice Fields as a Source of N₂O

The N₂O production is an integral part of the nitrogen transformation processes in wet soil fields. The biological processes of de-nitrification, nitrification, and dissimilatory nitrate reduction and assimilatory nitrate reduction, as well as the biological reaction of chemo-de-nitrification seems probable mechanisms of N₂O emission from soil. However, it has been established that de-nitrification and nitrification are the most important mechanism, with other process contributing very little to this phenomena.



MATERIALS AND METHODS

Study area

Two field experimental locations were selected from western Uttar Pradesh and the onsite experiments were carried out in rice farms in villages namely Chachula (Gautam Budhnagar) and Meerapur (Muzaffarnagar) of Indo-Gangetic-region of India. The sites are located in the Indo-Gangetic plain with coordinates 28°21'N and 77°35' E, 29°21'N and 78°00' E at an altitude of 191 m, 214 m respectively above mean sea level. The climate of the region is subtropical semi-arid and the mean maximum and minimum temperatures from July to October (rice season) are 34°C and 17°C season. For the study Rice (*Oryza sativa*) variety of Sughand-1121 and PR- 14 were planted and studied. Rice was transplanted in the flood field with 30 days old seedling. Irrigation in the rice fields was employed at 3-5 days intervals. The field based experiments were conducted in the year 2012.

Methane and nitrous oxide sample collections from rice fields

Methane and nitrous oxide emissions from paddy fields were monitored by employing fabricated acrylic chamber (length 50 cm, width 30 cm and height 100 cm) designed and fabricated in laboratory (Hutchinson and Mosier, 1981; Debnath *et al.*, 1996). Gas samples were drawn with 50 ml syringes with the help of a hypodermic needle (24 gauges). Methane and nitrous oxide emission rates from the paddy fields were determined at 0, 30, 60 min intervals (Kumar *et al.*, 2014) by measuring the changes of methane and nitrous oxide concentrations (the net change between greenhouse gas emission and sink) in the acrylic chamber.

Analysis of methane and nitrous oxide samples

Methane and nitrous oxide samples were collected and brought to the laboratory. Concentrations of CH₄ in the gas samples were analyzed by using Gas Chromatograph (GC) – model - SRI-8610C, USA fitted with a flame ionization detector (FID) with (Porapak-N) with stainless steel (2m) column and carrier gas hydrogen, nitrogen/argon and air and standard for CH₄ 5ppm was used. The N₂O concentration of the gas samples

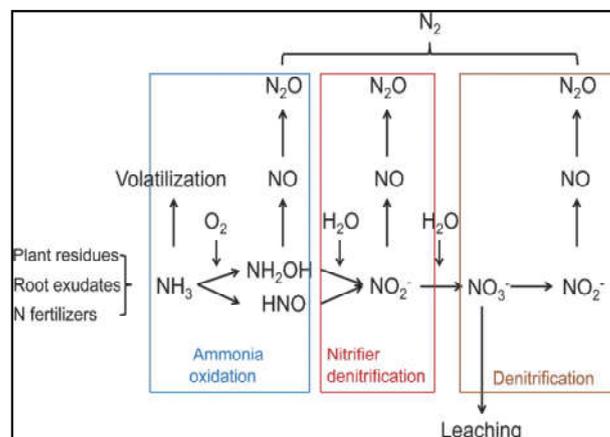


Fig.1. Nitrification and denitrification process in wet soils

Source: femsre.oxfordjournal.org & www.agnet.org

collected from the farms were also estimated by using GC. The N_2O samples were analyzed using electron capture detector (ECD) with 6'x 1/8'' Porapak-Q SS (2m) stainless steel column. The injector and detector temperatures were maintained at 50 °C, 120 °C, and 320 °C, respectively. Carrier gas was N_2 with a flow rate of 14 ml/min. The CH_4 and N_2O flux were calculated from the temporal increase of their mixing ratios inside the box.

Statistical analysis

Statistical comparisons of CH_4 and N_2O flux were performed with one-way analysis of variance (ANOVA). Analysis using ANOVA is performed to test whether the difference in means is statistically significant. The above statistical analyses were done using IBM SPSS Statistical Software (version 20.0; SPSS GGSIP University, Dwarka, New Delhi. In all cases, mean differences between group means were considered statistically significant if $p < 0.05$.

RESULTS AND DISCUSSION

The present study represents the actual field measurements of CH_4 emissions from Gautam Budhnagar district and Meerapur, Muzaffarnagar district in Indo-Gangetic region of India. Methane seasonal and N_2O seasonal flux values at each chamber site were found with changing trend for each set of flux measurements which were conducted simultaneously within a rice field during whole rice cropping season. The value of fluxes of CH_4 fluctuated between 0.348 and 1.343 $Kg ha^{-1} d^{-1}$ at chachula and 3.794 and 13.686 $Kg ha^{-1} d^{-1}$ at Meerapur during the rice season and no specific pattern was observed in any of the chamber of chachula (Fig.3) and Meerapur (Fig.4) of Indo-Gangetic region. It is understood that the anaerobic conditions are generally prerequisite for the formation of CH_4 in moist soil environment. Seasonal CH_4 emission variations were mainly controlled by various parameters depending on the weather conditions and soil



Fig.2. Close chamber technique employed in rice fields for CH_4 and N_2O gases sampling

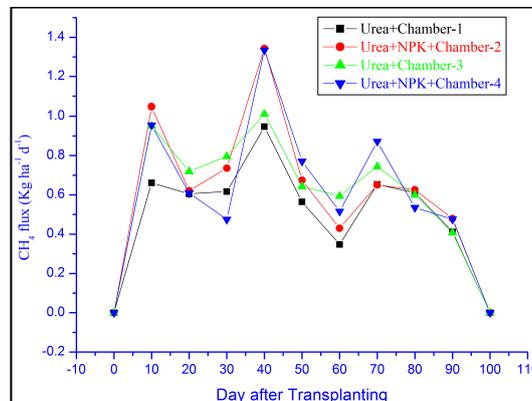


Fig.3. Variations of methane (CH_4) fluxes from rice-cropping system at Chachula

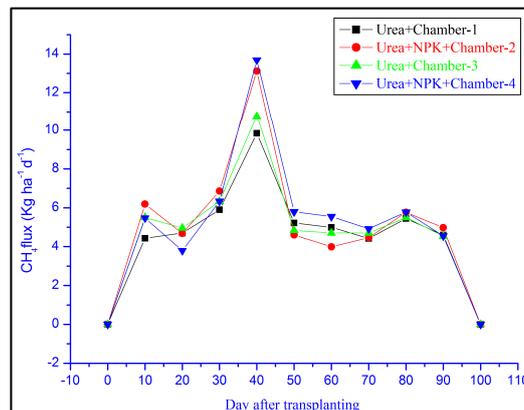


Fig.4. Variations of methane (CH_4) fluxes from rice-cropping system at Meerapur

characteristics these may be like temperature, redox potential (Eh), plant biomass, soil pH and Soil organic carbon (SOC) and moisture available in the paddy field. It may be noted that the higher CH₄ flux are observed in afternoon validating a major influence of soil temperature which play a vital role during the methanogenesis process.

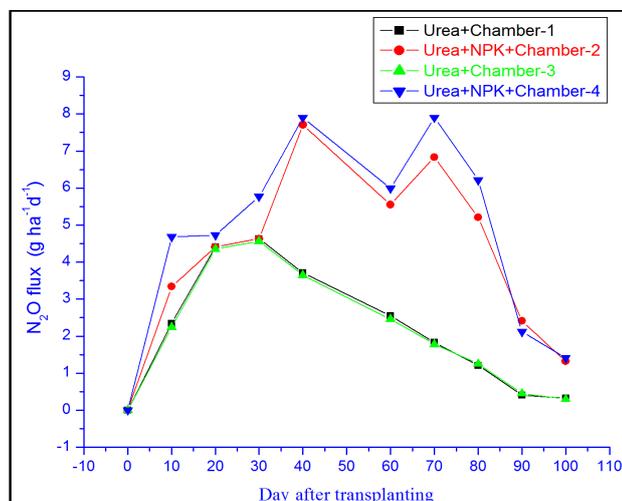


Fig.5. Variations of Nitrous oxide (N₂O) fluxes from rice-cropping system at Chachula

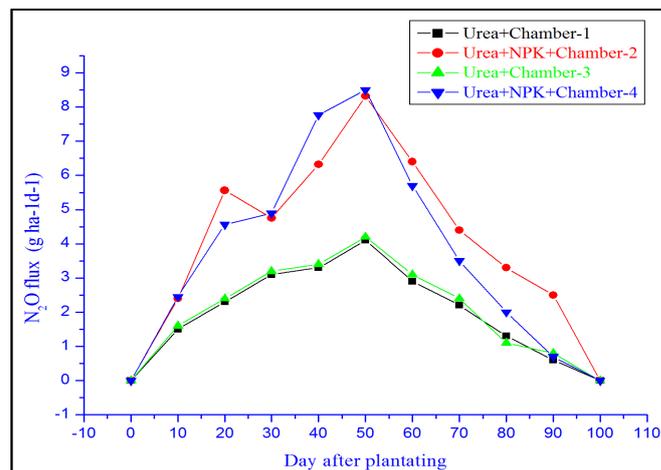


Fig.6. Variations of Nitrous oxide (N₂O) fluxes from rice-cropping system at Meerapur

The value of fluxes of N₂O a prominent greenhouse gas fluctuated between 0.30 to 7.9 gha⁻¹d⁻¹ at chachula and 0.60 to 8.5 gha⁻¹d⁻¹ at meerapur from the chambers which were fixed at the sites during the rice season. The values of CH₄ flux from Fig. 3 and Fig. 4 clearly suggest that flux has increased from tillering to panicle primordia initiation stage at both the

Table 1. One-way ANOVA test summary for CH₄ flux at A (Chachula) and B (Meerapur)

Groups	Count	Sum		Average		Variance	
		A	B	A	B	A	B
Column 1	9	5.418	49.536	0.602	5.504	0.028435	2.878192
Column 2	9	6.607	54.649	0.734111	6.072111	0.082652	7.817159
Column 3	9	6.464	51.821	0.718222	5.757889	0.034848	3.837273
Column 4	9	6.542	55.879	0.726889	6.208778	0.083094	8.438003

Table 2. One-way ANOVA test for CH₄ flux at A (Chachula) and B (Meerapur)

Source of Variation	SS		df	MS		F-values	
	A	B		A	B	A	B
Between Groups	0.10561	2.71043	3	0.035203	0.903477	0.614829	0.157327
Within Groups	1.832233	183.765	32	0.057257	5.742657		
Total	1.937844	186.4755	35				

Table 3. One-way ANOVA test summary for N₂O flux at A (Chachula) and B (Meerapur)

Groups	Count	Sum		Average		Variance	
		A	B	A	B	A	B
Column 1	9	21.41	21.3	2.378889	2.366667	2.597336	1.2175
Column 2	9	41.41	43.95	4.601111	4.883333	4.137336	3.9048
Column 3	9	21.07	22.2	2.341111	2.466667	2.484986	1.2775
Column 4	9	46.71	40.07	5.19	4.452222	5.11125	6.770069

Table 4. One-way ANOVA test for N₂O flux at A (Chachula) and B (Meerapur)

Source of Variation	SS		df	MS		F-values	
	A	B		A	B	A	B
Between Groups	59.42836	46.48887	3	19.80945	15.49629	5.529155	4.706588
Within Groups	114.6473	105.359	32	3.582727	3.292467		
Total	174.0756	151.8478	35				

The number of measurements per day was divided for morning and afternoon sessions. In morning hours the sampling was started from 9:00 am and the final reading was taken at 10:00 am. In afternoon the samples were collected after 2:00 pm keeping in mind the rise in temperature of the soil takes place in afternoon.

locations. The value has decreased afterwards i.e. from booting to harvest stage. On the other hand from fig. 5 and fig. 6 the values of N₂O flux show an increasing trend from tillering to heading/anthesis stage. The value has decreased afterwards in ripening stage i.e. from milky to maturity. It can be inferred from the results that the value of flux for N₂O has increased for

10 more days in comparison to CH₄ flux. Table 1 and Table 3 show the one-way ANOVA summary for CH₄ and N₂O flux at two different locations selected for the study. Also the results obtained on application of one-way ANOVA on CH₄ and N₂O flux due to variation in sampling locations are given in table 2 and table 4 respectively. From Table 4, it was understood that the variation in flux values for N₂O based on locations are significant at P<0.05.

Conclusion: The present investigation was carried out to find out the variation in CH₄ and N₂O greenhouse gases at two different locations of IGP region. CH₄ and N₂O emissions from rice-cropping system of western Uttar Pradesh show clear temporal and spatial variations. There is a commutation relation between methane and nitrous oxide emissions at both the locations of Chachula and Meerapur. In rice field soils sporadic watering can significantly reduce CH₄ emissions and increase N₂O emissions. However, the overall effect of green house can be reduced without affecting the rice yield. The periodic irrigation is a crucial and effective measure to reduce greenhouse gases from paddy fields. It can be seen from the fig. 3, fig. 4, fig. 5 and fig. 6 that the emissions in both the cases of CH₄ and N₂O are greatly influenced by the application of NPK blended with urea. The emissions of CH₄ as well as that of N₂O are increased with the application of commercial fertilizer in both the locations of Chachula and Meerapur from western Uttar Pradesh of Indo-Gangetic region. The microbial processes have also significant effects on CH₄ and N₂O emissions from rice fields and the emissions can be reduced by adopting intermittent wetting and drying of soils in paddy fields.

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