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# Identifying optimum rates of fertilizer nitrogen application to maximize economic return and minimize nitrous oxide emission from rice–wheat systems in the Indo-Gangetic Plains of India

Tek B. Sapkota<sup>a</sup>, Love K. Singh<sup>b</sup>, Arvind K. Yadav<sup>c</sup>, Arun Khatri-Chhetri<sup>d</sup>, Hanuman S. Jat<sup>e,f</sup>, Parbodh C. Sharma<sup>f</sup>, Mangi L. Jat<sup>g</sup> and Clare M. Stirling<sup>h</sup>

<sup>a</sup>Sustainable Intensification Program, International Maize and Wheat Improvement Centre (CIMMYT), El Batan, Mexico; <sup>b</sup>Borlaug Institute for South Asia (BISA)/CIMMYT, Ludhiana, India; <sup>c</sup>Department of Agronomy, Sri Karan Narendra Agricultural University, Jobner, India; <sup>d</sup>CGIAR Research Program on Climate Change, Agriculture and Food Security (CCAFS), Borlaug Institute for South Asia (BISA)/CIMMYT, New Delhi, India; <sup>e</sup>Sustainable Intensification Program, International Maize and Wheat Improvement Centre (CIMMYT), CSSRI, Karnal, India; <sup>f</sup>ICAR-Central Soil Salinity Research Institute (CSSRI), Karnal, India; <sup>g</sup>Sustainable Intensification Program, International Maize and Wheat Improvement Centre (CIMMYT), New Delhi, India; <sup>h</sup>Sustainable Intensification Program, International Maize and Wheat Improvement Centre (CIMMYT), World Agroforestry Centre (ICRAF), Nairobi, Kenya

## ABSTRACT

Rice–wheat (RW) cropping system in India is a major source of N<sub>2</sub>O emissions. In such system, defining N rates that deliver minimal N<sub>2</sub>O emissions and economically optimum yield would benefit both food production and the environment. We measured yield and N<sub>2</sub>O fluxes from RW systems in Northwest IGP under two tillage systems and five N rates (0, 75, 150, 225 and 300 kg N ha<sup>-1</sup>) for both rice and wheat using static chamber method. Seasonal pattern of N<sub>2</sub>O emission was mainly influenced by fertilizer and water application events with no significant effect of tillage systems. Mean annual N<sub>2</sub>O emission from RW system was 1.49 kg N ha<sup>-1</sup> in N75 plot and 2.97–3.04 in the plots receiving  $\geq 150$  kg N ha<sup>-1</sup>. On average, the yield-scaled N<sub>2</sub>O emissions of rice and wheat were 0.25 and 0.52 kg N<sub>2</sub>O–N mg<sup>-1</sup>, respectively. Our finding suggests that N rates between 120–200 kg N ha<sup>-1</sup> in rice and 50–185 kg ha<sup>-1</sup> in wheat provide the most economical returns and application rates beyond these ranges would be both economically and environmentally unsustainable. Within the range of N rate studied, fertilizer-induced N<sub>2</sub>O-EF for rice and wheat were 0.41% and 0.79%, respectively.

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## KEYWORDS

Nitrous oxide; emission factor; greenhouse gas; rice–wheat system

## Introduction

Nitrous oxide (N<sub>2</sub>O) plays important roles in influencing stratospheric chemistry and regional and global climate change (IPCC 2013). Although present in small quantities, N<sub>2</sub>O has a global warming potential approximately 265 time greater than that of CO<sub>2</sub> (Myhre et al. 2013) over a 100 years' time horizon. N<sub>2</sub>O is also responsible for depleting stratospheric ozone (Ravishankara et al. 2009). Therefore, developing and implementing methods to reduce N<sub>2</sub>O emissions from agricultural croplands is important. Agricultural soils account for approximately 60% global anthropogenic N<sub>2</sub>O emission (Foley et al. 2011), N fertilization in croplands being one of the major sources (Cole et al. 1997). Addition of N fertilizer (both synthetic and organic) to agricultural soils leads to N<sub>2</sub>O emissions,

**CONTACT** Tek B. Sapkota ✉ [t.sapkota@cgiar.org](mailto:t.sapkota@cgiar.org) International Maize and Wheat Improvement Centre (CIMMYT), NASC complex, New Delhi 110012, India

Supplemental data for this article can be accessed [here](#).

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predominantly by the microbial processes of nitrification (oxidation of ammonium to nitrate) and denitrification (reduction of nitrate to  $N_2$  with  $N_2O$  as intermediary product). Numerous studies have shown that  $N_2O$  emissions from agricultural soils are directly related to N application rates (Bouwman et al. 2002; Halvorson et al. 2011; Hoben et al. 2011). Therefore, farmers in many regions of the world are being incentivized through policies and supply chains to improve nutrient use efficiency (NUE) through better in-field management to protect water resources and minimize GHG emissions whilst sustaining food production.

The rice–wheat (RW) system covering 16 million ha of productive land in Asia (Pathak et al. 2002) is important for the food security of millions of people (Ladha et al. 2009). This production system is also a major consumer of fertilizer N in the region. The majority of rice and wheat farmers in the IGP and elsewhere in South Asia apply fertilizer N following blanket recommendations based upon crop response data averaged over large geographic areas. Given the wide spatial variability in indigenous nutrient supplying capacity of soils in different agro-ecologies, crop fertilization following such blanket recommendation results in under-fertilization in some cases and over-fertilization in others. Diagnostic surveys in the IGP revealed that farmers often apply greater than recommended rates of fertilizer N and P, but overlook the need to balance this with sufficient application of potassium and other secondary and micro-nutrients (Singh et al. 2005). Because the government of India (GoI) provides more subsidy on nitrogenous fertilizer than other nutrients, farmers often apply fertilizer N in doses even higher than the blanket recommendations. Such imbalances and inadequate use of nutrients reduces the NUE and profitability and may increase environmental risks associated with loss of unutilized nutrient through emissions or leaching (Sapkota et al. 2014b). At present, total fertilizer-N consumption in India is about 17 million tonnes (Tewatia and Chanda 2017), about 70% of which is consumed in cereal production (Bijay-Singh 2017). The necessity to increase food production to feed an increasing population means that the consumption of fertilizer-N will continue to grow in the future and is projected to be around 24 million tonnes by 2030 (Tewatia and Chanda 2017). Therefore, fertilizer-induced  $N_2O$  emission from agriculture is expected to increase further in India.

Fertilizer N amount, source, timing and placement can all influence  $N_2O$  flux from agricultural soils (Snyder et al. 2007). Stehfest and Bouwman (2006), in an extensive review of published studies, concluded that soil  $N_2O$  flux is best predicted by N application rate, N source, crop type, soil pH, soil texture, climate and soil organic matter (SOM). Among these, total input of N to the soil is considered one of the strongest predictors of  $N_2O$  emissions and this together with the emission factors (EF) are widely used to estimate  $N_2O$  emissions (Shcherbak et al. 2014). Through review of peer-reviewed articles and modeling, Albanito et al. (2017) reported mean  $N_2O$  EF as 1.2% for tropics and sub-tropics, which is within the uncertainty range of IPCC Tier 1 EF, i.e. 0.3–3% (De Klein et al. 2006). This means, for every 100 kg of N input, 1.2 kg of N in the form of  $N_2O$  is estimated to be emitted directly from the soil. However, results from a growing number of field experiments indicate that the fraction of applied N emitted as direct  $N_2O$  increases with increasing rate of N application (McSwiney and Robertson 2005; Ma et al. 2010; Hoben et al. 2011; Shcherbak et al. 2014; Millar et al. 2018). This is likely due to competition for available N between plant uptake and  $N_2O$  producing microbes; such that when crop demand is met more  $N_2O$  is produced per unit of additional N (Ma et al. 2010). Given this biological threshold, use of single EF across the fertilizer rates may underestimate fertilizer-induced  $N_2O$  emission where fertilizer N application is higher than crop demand.

According to India's second national communication to the United Nations Framework Convention on Climate Change (UNFCCC), fertilizer-induced field emission of  $N_2O$  in India was estimated to be 60 million tonnes  $CO_2e$  (GOI/MoEF 2012). A dearth of measurements under different crops, cropping systems and agro-ecology makes national and sub-national estimates highly uncertain. Analysis of yield and N input of the cropping systems in India during 1961–2009 demonstrates that crop N input (N rate) is increasing over time and corresponding NUE is decreasing (Bijay-Singh 2017). This indicates that substantial amounts of unused N under high N-input cropping systems may have been lost through direct and indirect emissions suggesting that fertilizer-induced  $N_2O$  EF may be much higher in these production systems. In the production systems such as RW system of

IGP where fertilizer N is often applied at a rate exceeding crop demand. In such systems defining N rates where yields are maximized and environmental harm are minimized could benefit both food production, human health and the environment. We therefore conducted a field study to quantify yield and N<sub>2</sub>O emissions under five N rates in RW systems of North-West IGP. The objectives of the study were to (i) identify a target range of N rates for RW system for optimal yields, profits and N<sub>2</sub>O emissions that are compatible with sustainable intensification and (ii) develop fertilizer-induced N<sub>2</sub>O EFs for intensive RW systems. This will potentially improve the national N<sub>2</sub>O inventory as RW system is one of the most extensive and input-intensive cropping systems in India.

## Materials and methods

### *Study site and experimental details*

The study was conducted in the research farm of Central Soil Salinity Research Institute (CSSRI) of Indian Council of Agricultural Research (ICAR) located in Karnal district of Haryana, India (29°70' N, 76°96'E.; 250 m above sea level). The experimental site is a reclaimed alkali loam soil and the field was under a continuous RW system for over half a century before establishment of the experimental platform. The soil characteristics of the site are given in Supplementary Table 1. The climate of the location is semi-arid with average annual rainfall of 700 mm, 75–80% received during June–September. The lowest temperature is observed during January (daily minimum ranged from 0 to 4°C) and the highest temperature is observed during June (daily maximum ranges from 40 to 44°C). Monthly rainfall distribution along with minimum and maximum temperature during the experimental seasons is presented in Supplementary Figure S1.

We started the experiment from the wheat season in 2014–2015 (Nov–March), continued through the rice season in 2015 (July–October) to the wheat season in 2015–2016. We evaluated five N rates (i.e. 0, 75, 150, 225 and 300 kg N ha<sup>-1</sup>) in both rice and wheat grown under two tillage systems (i.e. conventional tillage, CT and zero-tillage, ZT). Five N rates (sub-plot factors) were factorially combined with two tillage systems (main-plot factor) in split-plot design with three replications giving a total of 30 sub-plots of 10 by 10 m each.

### *Crop management*

Crop management including land preparation, variety, seed rate, sowing/planting, as well as management of water, nutrient and pest for rice and wheat are summarized in Table 1. Tillage operations in CT-based rice involved three passes of dry tillage with harrow to the depth of 15 cm, two passes of cultivator in ponded water (puddling) followed by planking (levelling). CT-based wheat received two passes of harrow, one pass of cultivator followed by one planking. In CT systems, rice seedlings (variety CSR-30) were raised using a seed rate of 12 kg ha<sup>-1</sup> and 30 days old seedlings were transplanted manually in random geometry with about 30 seedlings m<sup>-2</sup>. In this system, wheat seeds were broadcasted after all tillage operations followed by planking. In the zero-tillage system, both rice and wheat were seeded without any preparatory tillage at a row spacing of 22.5 cm using ZT seed-cum-fertilizer drill having inclined-rotary-plate seed metering systems. In ZT system, the seed rate was 25 and 100 kg ha<sup>-1</sup> for rice and wheat, respectively. Seeding depth was maintained at ~2 cm and 5 cm in rice and wheat crop, respectively using a depth-control wheel of the planter. The field remained fallow after harvesting of the first year wheat until establishment of rice in rainy season. Crops were fertilized by manually broadcasting fertilizer as per the treatment. Both rice and wheat received 60 kg P<sub>2</sub>O<sub>5</sub> and K<sub>2</sub>O ha<sup>-1</sup> through single super phosphate (SSP) and muriate of potash (MoP), respectively. N fertilization was done as per the treatments using Urea. Full amount of P and K and 33% of N was applied as basal at the time of seeding/transplanting and the remaining two-third of N was applied in two equal splits at 20–25 and 40–45 days after seeding/transplanting in rice and wheat.

**Table 1.** Details of tillage, crop establishment and residue management of rice and wheat under two tillage systems (main-plot factors).

Activity/Operations	CT		ZT	
	Rice	Wheat	Rice	Wheat
Cultivar	CSR-30	HD-2967	CSR-30	HD-2967
Field preparation	Harrowing (×3), cultivators (×2) for puddling followed by planking.	Harrowing (×2), cultivators (×1) followed by planking	ZT	ZT
Date of sowing/transplanting	30 July 2015	13 November 2014 18 November 2015	31 June 2015	13 November 2014 18 November 2015
Seed rate (kg ha <sup>-1</sup> )	12	150	25	100
Method	Transplanting in random geometry	Broadcasted in random geometry	Drill (line sowing)	Drill (line sowing)
Row spacing (cm)	Random	Random	20	20
Nutrient management	P <sub>2</sub> O <sub>5</sub> and K <sub>2</sub> O: 60 kg ha <sup>-1</sup> through SSP and MoP, respectively N: as per treatment	P <sub>2</sub> O <sub>5</sub> and K <sub>2</sub> O: 60 kg ha <sup>-1</sup> through SSP and MoP, respectively N: as per treatment	P <sub>2</sub> O <sub>5</sub> and K <sub>2</sub> O: 60 kg ha <sup>-1</sup> through SSP and MoP, respectively N: as per treatment	P <sub>2</sub> O <sub>5</sub> and K <sub>2</sub> O: 60 kg ha <sup>-1</sup> through SSP and MoP, respectively N: as per treatment
Water management	Continuous flooding of 5-cm for one month, then irrigation applied at appearance of hair-line crack	Four irrigation at CRI, tillering, flowering and grain filling	First irrigation at sowing. Second irrigation one week after sowing. Subsequent irrigations at appearance of hair-line crack	Four irrigation at CRI, tillering, flowering and grain filling
Weed management	Post-emergence: bispyribac sodium (25 ml a.i. ha <sup>-1</sup> )	Post-emergence: metsulfuron methyl (20 g a.i. ha <sup>-1</sup> )+ clodinafop (400 g a.i. ha <sup>-1</sup> )	Pre-sowing: glyphosate (1.25 g a.i. ha <sup>-1</sup> ) Pre-emergence: Pendimethalin (1.0 L a.i. ha <sup>-1</sup> ) Post-emergence: bispyribac sodium (25 g a.i. ha <sup>-1</sup> )	Pre-sowing: glyphosate (1.25 g a.i. ha <sup>-1</sup> ) Post-emergence: metsulfuron methyl (20 g a.i. ha <sup>-1</sup> )+ clodinafop (400 g a.i. ha <sup>-1</sup> )
Harvesting	26 October 2015	14 April 2015 09 April 2016	26 October 2015	14 April 2015 09 April 2016

During the rice cycle, CT plots were kept continuously flooded (5 cm standing water) for the initial one month and the subsequent irrigations were scheduled when soil matrix potential reaches around  $-40$  to  $-50$  kPa determined through a visual inspection of hairline cracks on the soil surface (Gathala et al. 2011). In ZT plots, the first irrigation was applied immediately after sowing. The second irrigation was given one week after seeding and subsequent irrigations were applied as described under CT system. Irrespective of the tillage systems, wheat received four irrigations (6–7 cm each) at 20–25, 45–50, 75–80 and 95–100 days after sowing. In ZT plots, weeds prior to seeding of rice and wheat were killed by pre-plant application of glyphosate but no herbicides were applied in CT plots before seeding. Weed management after seeding/transplanting in rice and wheat were done by using appropriate pre- and post-emergence herbicide as required (Table 1).

### **Grain and biomass yield estimation**

At maturity, crops were harvested manually from three  $1\text{-m}^2$  quadrats randomly selected within each plot for both rice and wheat. The plants within at least 0.5 m from the border were not considered for yield determination. The harvested crops were sun-dried and threshed to determine grain and straw yield. Grain and straw yield of rice and wheat were reported at 14% moisture content.

### **Collection of gas sample**

Gas samples were collected using two-part static chambers as described by Sapkota et al. (2014a). The base of the chamber (43 cm i.d.) made up of the galvanized steel was semi-permanently installed (12 cm of the base inserted into the soil keeping 17.5 cm above soil surface) in the plots keeping 5–10 plants inside the chamber. It consisted of a circular channel to hold the upper part of the chamber. At the time of sampling, the upper part of the chamber was placed over the base of the chamber giving a total headspace volume of 105.41 L. The circular channel was filled with water and vents were sealed with adhesive to make the assembly airtight. The chamber top was equipped with a battery-powered fan to facilitate mixing of the gas in chamber headspace. Gas sampling was commenced one day before seeding in each crop. Thereafter, gas samples were collected once a week and for five consecutive days after every N fertilization events. Gas samples were collected through a septum fitted on the side of the chamber using a 50 mL polypropylene disposable syringe with three-way luer lock. The gas (50 mL) in the syringe was injected into the pre-evacuated and labelled 30 mL vials, which ensured higher pressure inside the vial to avoid contamination from ambient air. At each sampling, gas samples were collected four times within a total chamber deployment period of 30 min at 10-min interval. Sampling was performed between 9.00 and 11.30 am when soil surface temperature is believed to be equal to the daily average (Sapkota et al. 2014b). Chamber temperature was also recorded during gas sampling event using a thermometer fitted on the chamber top to be used for flux calculation. Depth of flood water when present and plant volumes inside the chamber were determined and subtracted from chamber volume to calculate effective chamber volume.

### **Gas analysis and calculations**

Collected air samples were analyzed for  $\text{N}_2\text{O}$  using a Gas Chromatograph (GC) (model: Bruker 450) equipped with Electron Capture Detector (ECD) with the temperature settings of  $300^\circ\text{C}$ . Argon and 5% methane were used as carrier gases with a flow rate of  $60\text{ mL min}^{-1}$ .

Concentrations of gases were calculated by comparing relative peak areas against the curves prepared from known concentrations (0.5, 1 and 10 ppm) of standard gases from Linde Engineering India Pvt. Ltd. To address the issue of GC drift, GC was calibrated periodically using  $\text{N}_2\text{O}$  standards of known concentration. Gas concentration at each sampling period was converted into mole of gas by using ideal gas law as below.

$$PV = nRT \quad (1)$$

where  $P$  = pressure,  $V$  = volume,  $n$  = number of moles of gas,  $R$  = the gas law constant and  $T$  = temperature.

The mole unit of gas was then converted into weight of gas considering the molecular weight of a particular gas. The daily  $N_2O$  emission rate was calculated from the linear increase (slope) in  $N_2O$  concentration over time. The increase in gas concentration over time was carefully monitored and only the data-points where  $t$ -test of the slope of the regression were significant ( $p = 0.05$ ) and were included in cumulative seasonal emission calculations. The fluxes in between two sampling dates were estimated by linear interpolation. Seasonal cumulative  $N_2O$  emission was calculated from the sum of daily emission rates between planting to harvesting of the crop. The direct  $N_2O$  emission factor (EF%) induced by the N fertilizer was calculated as  $EF = 100 \times (E_F - E_0)/N$ , where  $E_F$  ( $kg\ N\ ha^{-1}$ ) is the seasonal cumulative  $N_2O$  flux from N-fertilizer treatment,  $E_0$  ( $kg\ N\ ha^{-1}$ ) is the seasonal cumulative  $N_2O$  flux for non-fertilized treatment, and  $N$  is the seasonal N fertilizer application rate ( $kg\ N\ ha^{-1}$ ). To evaluate the environmental relevance of  $N_2O$  emissions under different N fertilizer management practices, the seasonal cumulative  $N_2O$  emission were divided by the grain yield to calculate yield-scaled  $N_2O$  emission. Total annual grain yield, cumulative emission, emission intensity and EF were calculated using the data from 2015 to 2016 rice and wheat seasons.

### **Economic and statistical analyses**

The amount of N applied may vary depending on whether the priority is to increase crop productivity, reduce emissions or maximize the rate of return from the use of N fertilizer. The best-case scenario would be to maximize yield and return whilst minimizing emissions from crop cultivation (multi-objective target). This study estimated productivity, emissions and return in relation to N rate. Grain production was converted to a money value based on the Minimum Support Price (MSP) for the study area, Karnal District of Haryana, India. The MSP is an agriculture product price set by Food Corporation of India (FCI) to purchase directly from the farmer (<http://fci.gov.in>). This rate is to safeguard the farmer to a minimum profit for the harvest, if the open market price is less than the cost of cultivation. The commission for Agricultural Cost and Prices (CACP), under the Ministry of Agriculture and Farmers welfare determines the MSP of particular agricultural products based on cost of cultivation and market prices of inputs and outputs in a particular location (<https://cacp.dacnet.nic.in/>).

Prices of paddy and wheat straws were collected from IndiaMART (<https://dir.indiamart.com>) for the study sites. Both grain and straw prices of paddy and wheat were also verified with the farmers from the study district. This study estimated an economically feasible range of N use in rice and wheat crops. The economically feasible range of N use indicates that return from additional N use in the crop cultivation is greater than the additional cost of N. When additional cost of N use is greater than additional return, application of N is no more economically suitable. Price of N fertilizer was collected from the Indian Farmers Fertilizer Cooperative (IFFCO: [www.iffco.in](http://www.iffco.in)) for the study sites which was further verified with the farmers from the study district. The incremental benefits of N use were estimated for different levels N rate for both paddy and wheat cultivation using Equation (2).

$$\text{Incremental Benefit (IB)} = \text{Incremental Return} - \text{Incremental Cost of N use} \quad (2)$$

A functional form of return to N use and emissions intensity was estimated using a nonlinear regression model. We used quadratic function to estimate the N rate response to economic return. Economic return is directly related to N rate response to crop output (yield), which is normally nonlinear (quadratic). Quadratic function indicates that N-response to yield initially increases and after reaching biological optimum the response starts decreasing. All data were analyzed following an Analysis of Variance (ANOVA) for split-plot design using the Costat Software (CoHort 2012). The difference between treatment means was compared using a LSD test at  $P < 0.05$  (Gomez and Gomez 1984).

## Results

### Weather and environmental conditions

The growing season mean air temperature for 2014–2015 wheat, 2015 rice and 2015–2016 wheat were 16, 27 and 18°C, respectively (Supplementary Figure S1), which was similar to the long-term average of the location. Although minimum temperature was similar in both the years, maximum temperature particularly in April was higher in 2015–2016 wheat season than in 2014–2015 wheat season. The 2015 rice season received 370 mm rainfall, about 70% of the total annual precipitation (524 mm). The 2014–2015 wheat season received 247 mm rainfall which was about five times more than that received in 2015–2016 wheat season (56 mm) and 2.5 times more than the long-term average (110 mm).

### Crop production

The effect of tillage system was not significant for grain as well as biomass yield in both rice and wheat in both the years (Table 2). Except for grain yield in 2014–2015, N rate resulted in significant difference in grain and above-ground biomass yield of rice and wheat in both the years (Table 2, Figure 1). There was no significant interaction between tillage and N rate. Therefore, we report grain and straw yields of each crop averaged over two tillage treatments. Figure 1 shows the grain and straw yield of rice and wheat receiving different N rates averaged over the two tillage systems. Grain yield and above ground biomass were always significantly lower in control (zero N) plots compared to plots where N fertilizer was applied. However, no significant effect of N was observed on grain, straw and aboveground biomass yield for N rate of 150 kg ha<sup>-1</sup> and above in both the crops in all years (Figure 1).

### Seasonal trend of N<sub>2</sub>O emission

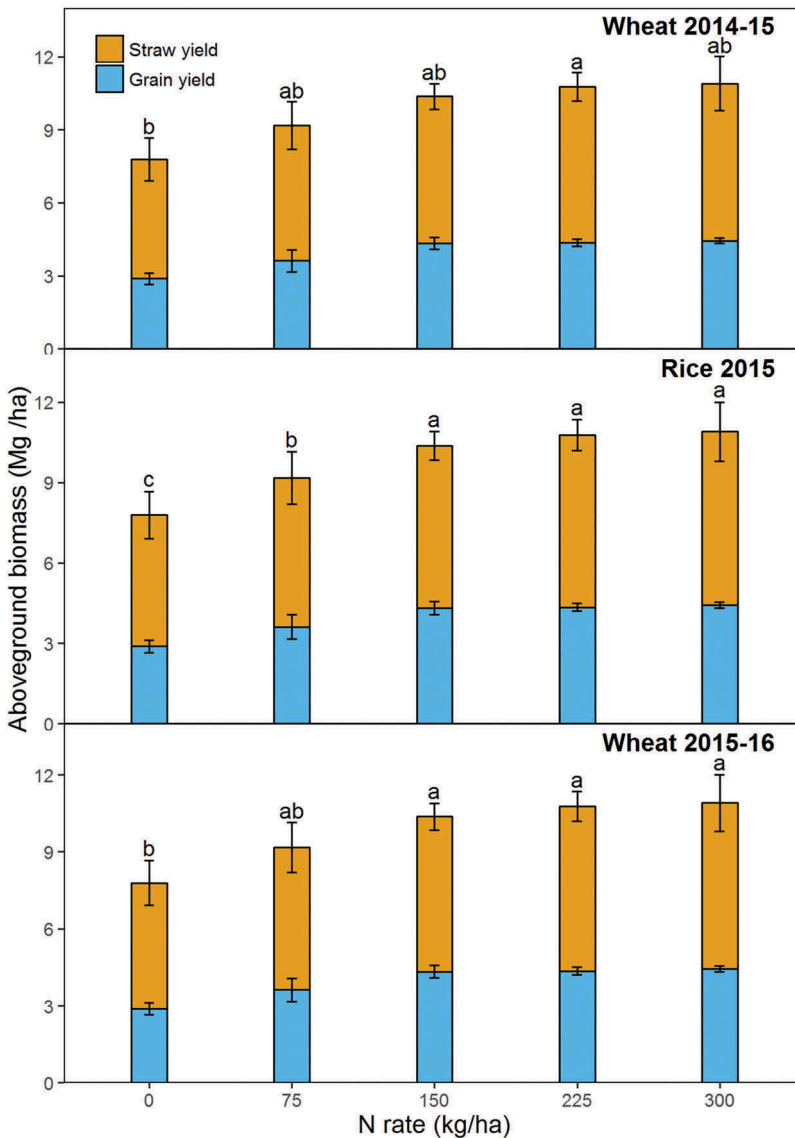
Nitrogen fertilization rates clearly influenced the daily soil N<sub>2</sub>O emissions during the wheat season in both years whereas no effect of tillage system was evident (Supplementary Figures S2 and S4). In

**Table 2.** Significance of effects of tillage systems, N rate (N) and their interactions on grain yield, total biomass yield, cumulative N<sub>2</sub>O emission, N<sub>2</sub>O emission intensity and N<sub>2</sub>O emission factor as resulting from analysis of variance (ANOVA).

Source of variation	df	Grain yield (mg ha <sup>-1</sup> )	Total biomass yield (mg ha <sup>-1</sup> )	N <sub>2</sub> O emission (kg ha <sup>-1</sup> )	N <sub>2</sub> O emission intensity (kg CO <sub>2</sub> e/mg grain)	N <sub>2</sub> O EF (%)
Wheat 2014–2015						
Tillage (T)	1	ns	ns	ns	ns	ns
N rate (N)	4	ns	**	***	ns	ns
T × N	4	ns	ns	ns	ns	ns
Rice 2015						
Tillage (T)	1	ns	ns	ns	ns	*
N rate (N)	4	**	**	***	**	ns
T × N	4	ns	ns	ns	ns	ns
Wheat 2015–2016						
Tillage (T)	1	ns	ns	ns	ns	ns
N rate (N)	4	**	***	***	***	*
T × N	4	ns	ns	ns	ns	ns
Rice–Wheat system						
Tillage (T)	1	ns	Ns	ns	ns	ns
N rate (N)	4	**	***	***	***	**
T × N	4	ns	ns	ns	ns	ns

\*, \*\* and \*\*\* are significant at 0.05, 0.01 and 0.001 probability level, respectively. ns is non-significant.





**Figure 1.** Rice and wheat grain and straw yield at maturity under different N rates (averaged over two tillage systems and three replications) during the experimental period of 2014–2016. The straw yield is stacked over grain yield to show total biomass yield (grain and straw). In each panel, the bars bearing different lowercase letters are significantly different from each other at  $P < 0.05$ . Error bars (standard error of the means) for grain and straw yield are separately provided.

general, daily soil  $N_2O$  emissions from the unfertilized control plots were lower than the N fertilized plots. Among the N fertilized plots, daily  $N_2O$  emission were lower in plots fertilized with  $75 \text{ kg N ha}^{-1}$  than those fertilizers at higher N rates, i.e.  $150$ ,  $225$  and  $300 \text{ kg N ha}^{-1}$ . The trend of  $N_2O$  emission was similar among the plots receiving  $150$ – $300 \text{ kg N ha}^{-1}$ . During both wheat seasons daily  $N_2O$  emissions from N fertilized plots increased immediately after sowing and after each fertilization event reaching to a maximum of  $14.67 \text{ mg N}_2\text{O-N m}^{-2} \text{ day}^{-1}$  in 2014–2015 wheat season and  $24 \text{ mg N}_2\text{O-N m}^{-2} \text{ day}^{-1}$  in 2015–2016 wheat season. The peak  $N_2O$  emission was observed 2–3 days after each fertilization event (Supplementary Figures S2 and S4). Although the  $N_2O$  emission trend was similar under CT and ZT plots, slightly higher  $N_2O$  emissions were recorded under ZT than CT plots across all fertilization treatments after the last fertilizer event in both the years.

In the rice-growing season, both tillage systems as well as N fertilization rates affected the trend of N<sub>2</sub>O emission. Irrespective of the tillage systems, daily emissions of N<sub>2</sub>O were smaller in the control plot than the N fertilized plots. The plots receiving 75 kg N ha<sup>-1</sup> also recorded lower daily and seasonal N<sub>2</sub>O emission than the plots receiving higher N rates, irrespective of the tillage system. The daily N<sub>2</sub>O emission from N fertilized plots during rice growing season ranged from 0 to 12.78 mg N<sub>2</sub>O–N m<sup>-2</sup> day<sup>-1</sup> (mean: 1.23 mg N<sub>2</sub>O–N m<sup>-2</sup> day<sup>-1</sup>) in CT plots and 0 to 22.39 mg N<sub>2</sub>O–N m<sup>-2</sup> day<sup>-1</sup> (mean: 2.31 mg N<sub>2</sub>O–N m<sup>-2</sup> day<sup>-1</sup>) in ZT plot. In ZT plots, N<sub>2</sub>O emissions were observed immediately after the first N fertilization whereas in CT plots no N<sub>2</sub>O emission was observed after the first dose of N application (Supplementary Figure S3). The magnitude of daily N<sub>2</sub>O emission in CT plots were smaller than those in ZT plots even after second dose of N fertilization. However, pronounced N<sub>2</sub>O peaks appeared following third dose of N application, particularly from the plots with higher N rate, i. e. 225 and 300 kg N ha<sup>-1</sup>. In ZT system, all the plots receiving 150 kg N ha<sup>-1</sup> or more recorded much higher daily N<sub>2</sub>O emission than other plots after second and third fertilization event.

### Cumulative N<sub>2</sub>O emission

Nitrogen fertilization rates had significant effect on cumulative N<sub>2</sub>O emissions in both rice and wheat growing seasons, whereas the effect of tillage systems was not significant. Similarly, tillage by N rate interaction effect was also not significant for soil N<sub>2</sub>O emissions from both rice and wheat in both the years (Table 2). Therefore, only the mean effect of N rates averaged over tillage systems are presented in Table 3.

**Table 3.** Seasonal and annual cumulative N<sub>2</sub>O fluxes, grain yield, emission intensity and N<sub>2</sub>O emission factor under different nitrogen rates from rice, wheat and rice–wheat. Within each crop and cropping system, means in columns bearing same lowercase letter are significantly different from each other based LSD test ( $p < 0.05$ ). Values are presented as means  $\pm$  SEM (standard error of the mean).

N rates (kg ha <sup>-1</sup> )	Cumulative N <sub>2</sub> O emission (kg N ha <sup>-1</sup> )	Grain yield (mg ha <sup>-1</sup> )	Emission intensity (kg N <sub>2</sub> O–N mg <sup>-1</sup> )	N <sub>2</sub> O emission factor (%)
2014–2015 Wheat				
0	0.88 $\pm$ 0.13 c	2.10 $\pm$ 0.36	0.54 $\pm$ 0.20 b	
75	1.48 $\pm$ 0.23 b	2.54 $\pm$ 0.45	0.72 $\pm$ 0.18 ab	0.81 $\pm$ 0.30
150	2.22 $\pm$ 0.15 a	2.84 $\pm$ 0.32	0.88 $\pm$ 0.15 a	0.84 $\pm$ 0.12
225	2.38 $\pm$ 0.07 a	3.76 $\pm$ 0.23	0.65 $\pm$ 0.05 ab	0.67 $\pm$ 0.08
300	2.40 $\pm$ 0.12 a	3.27 $\pm$ 0.51	0.88 $\pm$ 0.18 a	0.51 $\pm$ 0.05
2015 Rice				
0	0.28 $\pm$ 0.06 c	2.05 $\pm$ 0.05	0.14 $\pm$ 0.03 b	
75	0.62 $\pm$ 0.07 b	3.51 $\pm$ 0.08	0.18 $\pm$ 0.02 b	0.45 $\pm$ 0.12
150	1.15 $\pm$ 0.10 a	3.73 $\pm$ 0.05	0.31 $\pm$ 0.03 a	0.54 $\pm$ 0.09
225	1.14 $\pm$ 0.13 a	3.72 $\pm$ 0.07	0.31 $\pm$ 0.04 a	0.38 $\pm$ 0.06
300	1.16 $\pm$ 0.16 a	3.57 $\pm$ 0.06	0.33 $\pm$ 0.05 a	0.29 $\pm$ 0.06
2015–2016 Wheat				
0	0.09 $\pm$ 0.01 c	2.89 $\pm$ 0.22	0.03 $\pm$ 0.01 c	
75	0.87 $\pm$ 0.09 b	3.62 $\pm$ 0.41	0.25 $\pm$ 0.03 b	1.04 $\pm$ 0.11 a
150	1.83 $\pm$ 0.18 a	4.33 $\pm$ 0.22	0.42 $\pm$ 0.04 a	1.09 $\pm$ 0.09 a
225	1.89 $\pm$ 0.18 a	4.35 $\pm$ 0.14	0.44 $\pm$ 0.05 a	0.80 $\pm$ 0.08 ab
300	1.87 $\pm$ 0.28 a	4.44 $\pm$ 0.10	0.43 $\pm$ 0.07 a	0.59 $\pm$ 0.09 b
2015–2016 Rice–Wheat System				
0	0.37 $\pm$ 0.07 c	4.94 $\pm$ 0.17	0.07 $\pm$ 0.01 c	
75	1.49 $\pm$ 0.13 b	7.13 $\pm$ 0.41	0.21 $\pm$ 0.02 b	0.75 $\pm$ 0.08 ab
150	2.97 $\pm$ 0.24 a	8.06 $\pm$ 0.24	0.37 $\pm$ 0.03 a	0.81 $\pm$ 0.08 a
225	3.04 $\pm$ 0.22 a	8.07 $\pm$ 0.17	0.38 $\pm$ 0.03 a	0.59 $\pm$ 0.05 bc
300	3.02 $\pm$ 0.44 a	8.00 $\pm$ 0.14	0.38 $\pm$ 0.06 a	0.44 $\pm$ 0.08 c

The seasonal cumulative  $\text{N}_2\text{O}$  emission was significantly lower in control plot followed by N75, in which cumulative  $\text{N}_2\text{O}$  emission was significantly higher than control but significantly lower than the plots receiving higher N rates (i.e. N150, N225 and N300) in both the years (Table 3). The seasonal cumulative  $\text{N}_2\text{O}$  emission during the rice growing season ranged from 0.097 to 0.52 kg N ha<sup>-1</sup> (mean value: 0.28 kg N ha<sup>-1</sup>) for control plot. In the plots receiving N fertilization, average seasonal cumulative  $\text{N}_2\text{O}$  emission ranged from 0.62 kg N ha<sup>-1</sup> (in N75 plot) to 1.16 kg N ha<sup>-1</sup> (in N300 plot). Total cumulative  $\text{N}_2\text{O}$  emission from N75 plot in this season was significantly higher than the control but significantly lower than the plots receiving higher N rates. The plots receiving 150–300 kg N ha<sup>-1</sup> recorded the highest cumulative  $\text{N}_2\text{O}$  emission in this season (Table 3).

Seasonal cumulative emission from control plot in the wheat growing season ranged from 0.33 to 1.23 kg N ha<sup>-1</sup> (mean value: 0.88 kg h ha<sup>-1</sup>) during first year and 0.05 to 0.13 kg N ha<sup>-1</sup> (mean value: 0.09 kg N ha<sup>-1</sup>) during second year. In N75 plot, the cumulative  $\text{N}_2\text{O}$  emission for wheat season ranged from 0.99 to 2.33 kg N ha<sup>-1</sup> (mean value: 1.48 kg N ha<sup>-1</sup>) during first year and 0.58 to 1.14 kg N ha<sup>-1</sup> (mean value: 0.87 kg N ha<sup>-1</sup>) during second year (Table 3). N150, N225 and N300 plots had similar cumulative  $\text{N}_2\text{O}$  emission during both wheat growing season which ranged from 1.40 to 3.2 kg N ha<sup>-1</sup> (mean value: 2.10 kg N ha<sup>-1</sup>).

Annual  $\text{N}_2\text{O}$  emission (emission from entire crop year consisting of 2015 rice and 2015–2016 wheat season) ranged from 0.17 to 0.66 kg N ha<sup>-1</sup> (mean value: 0.37 kg N ha<sup>-1</sup>) for control which was significantly lower than the annual  $\text{N}_2\text{O}$  emission from other treatment (Table 3). Cumulative  $\text{N}_2\text{O}$  emission from annual RW rotation from N75 plot ranged from 1.08 to 1.88 kg N ha<sup>-1</sup> (mean value: 1.49 kg N ha<sup>-1</sup>), which was significantly higher than that of control plot but significantly lower than the plots receiving higher N rates (Table 3). All plots receiving N rates higher than 150 kg ha<sup>-1</sup> (i.e. N150, N225 and N300) resulted in similar cumulative emissions.

### **Yield-scaled $\text{N}_2\text{O}$ emission**

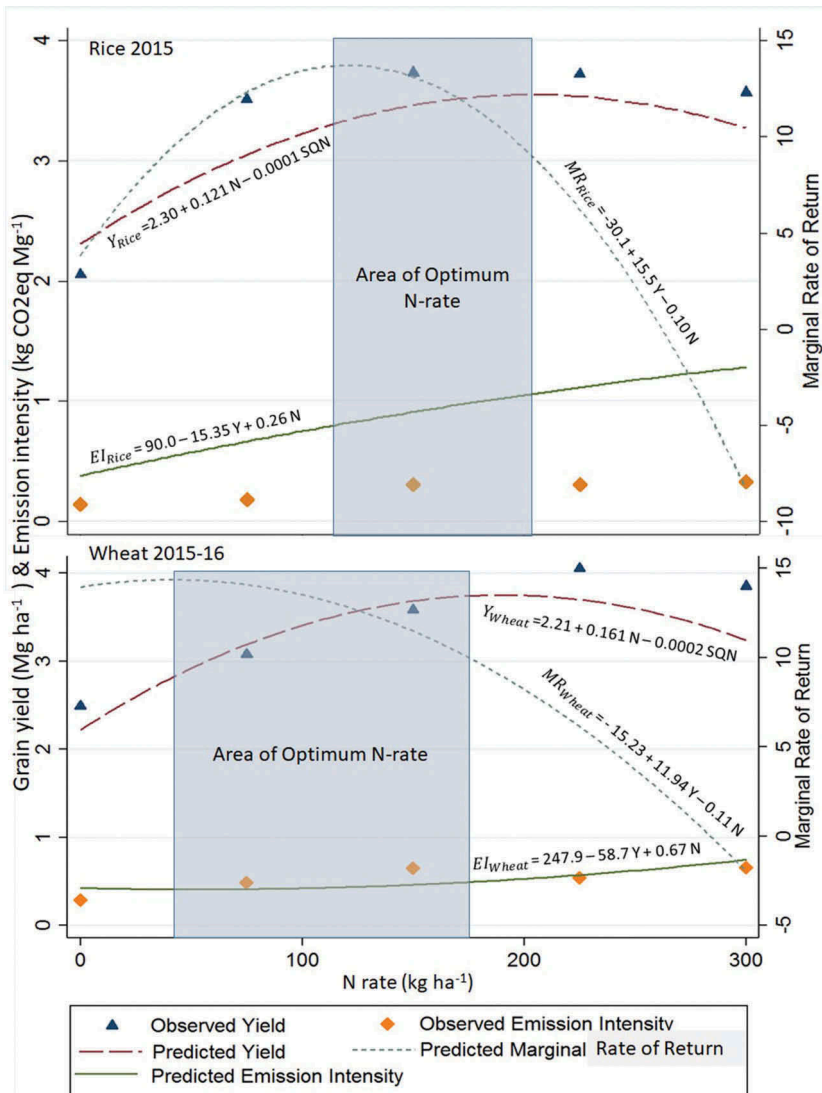
Yield-scaled  $\text{N}_2\text{O}$  emission (emission intensity) of rice and wheat in the studied year is presented in Table 3. Irrespective of the crops and year,  $\text{N}_2\text{O}$  emission intensity was higher in all plots receiving N rate 150 kg ha<sup>-1</sup> or more. The plots receiving 75 kg N ha<sup>-1</sup> resulted into significantly lower  $\text{N}_2\text{O}$  emission intensity than other N fertilized plots but significantly higher than the control plot (Table 3). Emission intensity of rice ranged from 0.04–0.55 kg  $\text{N}_2\text{O}$ -N mg<sup>-1</sup> whereas that of wheat ranged from 0.25–1.61 kg  $\text{N}_2\text{O}$ -N mg<sup>-1</sup> during 2014–2015 growing season and 0.02–0.77 kg  $\text{N}_2\text{O}$ -N mg<sup>-1</sup> during 2015–2016 growing season. As in rice and wheat season,  $\text{N}_2\text{O}$  emission intensity of RW rotation was intermediate in N75, control plots having significantly lower and the plots receiving  $\geq 150$  kg N ha<sup>-1</sup> having significantly higher emission intensity (Table 3).

### **Soil $\text{N}_2\text{O}$ emission factor (EF)**

Irrespective of treatments, EF for rice ranged from 0.08% to 0.91% whereas that for wheat range from 0.14% to 2.12% in 2014–2015 growing season and 0.435 to 1.41% in 2015–2016 growing season. Annual  $\text{N}_2\text{O}$  EF from RW system ranged from 0.26% to 1.08% across different N rates. The effect of tillage system and tillage by N rates interaction were not significant for seasonal and annual  $\text{N}_2\text{O}$  EF (Table 2). Main effect of N rates on  $\text{N}_2\text{O}$  EF was significant during 2015–2016 wheat (seasonal) as well as RW (annual) rotation (Tables 2 and 3). Overall,  $\text{N}_2\text{O}$  EF was higher with smaller N rates and *vice-versa*.

### **Optimum N rate**

A regression analysis was used to identify the relationship between marginal rate of return, N rate, and emissions intensity in both rice and wheat crops. This analysis estimated how N rate and emissions intensity behave with total economic return in both crops. Results of quadratic function showed that the N rate and emissions intensity in both rice and wheat crops were non-linearly



**Figure 2.** Observed and predicted grain yield, N<sub>2</sub>O emission intensity and predicted marginal rate of return under different N rates in rice (upper panel) and wheat (lower panel), 2015–2016.

related to total economic returns (Supplementary Table 2). The total economic return increased up to certain level of N-rate and emissions intensity, then started to decrease with increase in N rate (Square N Rate) and emissions intensity (Square Emissions Intensity) in both crops. Figure 2 indicates an area of ‘optimum’ N rate for rice (upper panel) and wheat (lower panel) crops. The optimum N ranges from the point of diminishing marginal rate of return to the point where yield stop increasing. The optimum rate of N fertilizer application in rice ranges from 120 to 200 kg ha<sup>-1</sup>. Similarly, the optimum rate of N fertilizer application in wheat ranges from 50 to 185 kg ha<sup>-1</sup>.

## Discussion

### *Seasonal N<sub>2</sub>O emission trend*

In both the crops, the fluxes of N<sub>2</sub>O were highly variable and demonstrated strong association with fertilization events. Although the mechanism of N<sub>2</sub>O generation and consumption in the soil is highly complex, it can be speculated that higher soil N content after urea application may have induced nitrification and denitrification-led N<sub>2</sub>O emission. Further, each fertilizer application event was followed by irrigation that may have triggered the activity of nitrifying and denitrifying microbes, which resulted in greater fluxes of N<sub>2</sub>O following fertilization event (Pathak et al. 2002; Sapkota et al. 2015).

The effect of tillage system on the seasonal trend of N<sub>2</sub>O emission was evident only in rice (Supplementary Figure S3) not in wheat seasons (Supplementary Figures S2 and S4). In ZT rice, N<sub>2</sub>O emission was observed right after basal fertilizer application though in smaller rate (Supplementary Figure S3 upper panel). In CT rice, on the other hand, the magnitude of N<sub>2</sub>O emission was smaller even after second fertilizer application probably because anaerobic conditions in the field following conventional tillage and flooding in this system might have suppressed the nitrification and hence associated N<sub>2</sub>O generation (Pandey et al. 2012; Butterbach-Bahl et al. 2013).

### *Cumulative N<sub>2</sub>O emission from rice and wheat crops*

N<sub>2</sub>O fluxes from rice and wheat fields have been measured in different countries and also in different cropping systems and different agro-ecologies in India (Pathak et al. 2002; Malla et al. 2005; Bhattacharyya et al. 2012; Pandey et al. 2012; Sapkota et al. 2015). Seasonal cumulative N<sub>2</sub>O emissions reported in the present study (0.58–3.23 and 0.38–1.93 kg N<sub>2</sub>O–N ha<sup>-1</sup> for wheat and rice season, respectively) are within the reported range in the previous literature. Relatively higher seasonal emission in 2014–2015 wheat season than 2015–2016 wheat season (Table 3) was probably due to evenly distributed rainfall during 2014–2015 wheat season (Supplementary Figure S1) resulting in soil conditions conducive (i.e. moist but not water-logged condition) to N<sub>2</sub>O emissions. Friedl et al. (2016) demonstrated through soil incubation study that denitrification-led N<sub>2</sub>O emission is higher in soil with 80% water-filled pore space than in 100% saturated soil. This is also evident from the higher background emissions (i.e. N<sub>2</sub>O emission from zero-N treatment) in the 2014–2015 wheat season than in any other crop seasons studied (Table 3). The background emission in our study ranged from 0.33–1.23, 0.09–0.52 and 0.07–0.13 kg N<sub>2</sub>O–N ha<sup>-1</sup> during first-year wheat, rice and second-year wheat, respectively (means shown in Table 3). In this study, background N<sub>2</sub>O emission accounted for 5–41% of total in fertilized treatments, which is comparable to the values (26–30%) reported by Gu et al. (2009). In terms of magnitude, background emission in this study was slightly smaller than that from rice–rapeseed systems in China, i.e. 0.66 kg N ha<sup>-1</sup> (Zhou et al. 2015). The differences in background emission can be attributed to different climatic conditions such as semi-arid climatic condition and soil properties such as lower soil total nitrogen content and low soil organic carbon content in our site. Such climate and soil conditions strongly affect the background emission of N<sub>2</sub>O (Gu et al. 2009).

Annual N<sub>2</sub>O emission from RW system in the fertilized treatments ranged from 1.07 to 5.17 (mean = 2.18) kg N<sub>2</sub>O–N ha<sup>-1</sup>. The annual N<sub>2</sub>O emission from RW system in N75 treatment in our study (1.49 kg N<sub>2</sub>O–N) was similar to the values (1.42 kg N<sub>2</sub>O–N) reported by Malla et al. (2005) from IGP with application of 120 kg N ha<sup>-1</sup> in both rice and wheat, whereas emission from the plots receiving higher rates of N were much higher (Table 3). On an average, rice and wheat contributed 40% and 60% of annual N<sub>2</sub>O emission, respectively. The contribution of rice and wheat to total annual N<sub>2</sub>O emission was respectively 36% and 64% in CT system and 42% and 58% in ZT system.

## ***N<sub>2</sub>O emission factor***

The N<sub>2</sub>O EF, the percentage of fertilizer N applied that is transformed into fertilizer-induced emission, was determined individually for the three crop seasons studied, i.e. wheat 2014–2015, rice 2015 and wheat 2015–2016. Annual N<sub>2</sub>O EF represents the data from the entire crop year consisting of 2015 rice and 2015–2016 wheat season. The fertilizer-induced N<sub>2</sub>O EF in our study (Table 3; 0.23–0.54% for rice and 0.59–1.09% for wheat) was on slightly lower side as compared to 1% proposed by IPCC (IPCC 2007) and also reported by Albanito et al. (2017) through a review and modelling of published EF from tropical and sub-tropical agricultural systems. As soil water can directly and indirectly influence N<sub>2</sub>O emission, prolonged dry period in our wheat field induced by semi-arid climatic condition (Supplementary Figure 1) coupled with limited number of irrigations might have suppressed nitrification-denitrification-induced N<sub>2</sub>O emission in our study. N<sub>2</sub>O EF in our study was smaller for rice which was comparable to 0.34% reported by Kumar et al. (2002) from the RW systems of IGP. EFs in our study did not increase with increasing N rate, contrary to a recent study (e.g. Shcherbak et al. 2014) who reported increased N<sub>2</sub>O-EF with increasing N rate. Given that N<sub>2</sub>O emission from soil is controlled by a multitude of soil and climatic variables and controlled by management factors, it is difficult to derive a general pattern of N<sub>2</sub>O dependency on N input. For example, Kim et al. (2013) examined the dependency of N<sub>2</sub>O emission on N input using published dataset and reported that N<sub>2</sub>O-EF remains constant or increase or decrease non-linearly with changing N rates.

We speculate that higher soil pH and low soil organic carbon in our study site (i.e. pH = 8 SOC = 0.56%; Table 1) are the main factors for absence of response of N<sub>2</sub>O emission at higher N rates. For example, Wang et al. (2017), through a global meta-analysis of 1,104 field measurements, reported that N<sub>2</sub>O emission in acidic soils is more sensitive to fertilization rate than that in alkaline soils. Further, in our C limited soil, increasing N rate above certain amount might have increase soil N beyond the capacity of soil microbes to take up and utilize thereby slowing down the N<sub>2</sub>O production rate and finally reaching a steady-state as hypothesized by Kim et al. (2013). Even in the meta-analysis of Shcherbak et al. (2014), higher N<sub>2</sub>O EF with higher N rates were evident only in the soils with carbon content >1.5% and pH<7. Although the apparent causality between soil pH and N<sub>2</sub>O emission has been studied previously, the influencing mechanisms of soil pH on N<sub>2</sub>O emission have not been completely understood. For example, soil liming in laboratory study suppressed N<sub>2</sub>O emission (Shaaban et al. 2015) and soil acidification through intensive fertilization significantly enhanced N<sub>2</sub>O emissions (Raut et al. 2012). It may be possible that lower soil pH may cause malfunction of N<sub>2</sub>O reductase enzyme, leading to higher N<sub>2</sub>O/N<sub>2</sub> ratio during denitrification (Bakken et al. 2012).

## ***Optimization of N fertilizer***

An ideal fertilizer rate for any crop would be the one that promotes the dual goals of high crop yield and low N<sub>2</sub>O flux (Sapkota et al. 2018). Although, the control treatment resulted in the lowest yield-scaled N<sub>2</sub>O emission (Table 3), it should be noted that for a N rate to be financially viable, it must produce sufficient crop yield. Both rice and wheat yields increased until N rate reached to 225 kg N ha<sup>-1</sup> (Figure 2), the rate of yield increment slowed down above these N rates in both the crops. The optimum rate of N fertilizer application in rice and wheat ranged from 120–200 to 50–185 kg N ha<sup>-1</sup>, respectively (Figure 2). This optimum range for N in both crop includes a range between maximum rate of marginal return and maximum yield with positive marginal rate of return. Therefore, application of N fertilizer above these ranges would not be financially viable or environmentally suitable.

## ***Conclusion***

This study provides the 'optimum' range for fertilizer N rate for rice and wheat to maximize crop yield and economic benefit and to minimize N<sub>2</sub>O emission. Overall, fertilizer-induced N<sub>2</sub>O-EF in our study

was about 0.42% in rice and 0.8% for wheat. Alkaline soil with low organic carbon in the experimental site might be responsible for reducing fertilizer-induced N<sub>2</sub>O emission in our study, particularly under high N application rate. Based on this analysis, N rate of 120–200 kg N ha<sup>-1</sup> for rice and 50–185 kg N ha<sup>-1</sup> for wheat is agronomically productive, economically viable and environmentally sustainable for the RW system of North-West Indo-Gangetic Plains.

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## Disclosure statement

No potential conflict of interest was reported by the authors.

## ORCID

Tek B. Sapkota  <http://orcid.org/0000-0001-5311-0586>

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