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ORIGINAL ARTICLE

Nitrous oxide and nitric oxide emissions and nitrogen use efficiency as affected by nitrogen placement in lowland rice fields

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Abstract Urea deep placement (UDP) has demonstrated its benefits of saving N fertilizer and increasing nitrogen use efficiency (NUE) and grain yields. However, studies on its environmental impacts, particularly on nitrous oxide (N_2O) and nitric oxide (NO), are limited. We conducted multi-location field experiments in Bangladesh to determine the effects of UDP versus broadcast prilled urea (PU) on N_2O and NO emissions, NUE, and rice yields. N_2O and NO emissions were measured from three N fertilizer treatments—no N, UDP, and PU—using automated gas sampling and analysis systems continuously for two rice-growing seasons—*Aus* (May–August) and *Aman* (August–December). Fertilizer-induced peaks in N_2O emissions were observed after broadcast application of PU but were rarely observed after UDP. Total seasonal N_2O and NO emissions, yield-scaled emissions, and fertilizer-induced emissions were affected by fertilizer treatments and sites. Though nitrogen fertilizer increased emissions significantly over the control, emissions resulting from UDP and PU were similar. Effects of N placement on grain yields and NUE were site- and season-specific. Of the N placement methods, UDP increased grain yields by 13% ($p < 0.05$) during the *Aman* season and gave similar yields in spite of lower N application during the *Aus* season. UDP increased N recovery from 25 and 16% of broadcast PU to 61 and 73% during the *Aus* and the *Aman* seasons, respectively in one site, but was similar in another site. On the other hand, alternate wetting and drying irrigation reduced grain yield and N recovery at the BRRI site during the *Aman* season.

Keywords Nitric oxide · Nitrogen use efficiency · Nitrous oxide · Greenhouse gas emissions · Urea deep placement · Rice

Introduction

Nitrous oxide (N_2O) is a major greenhouse gas (GHG) and 265 times more potent in terms of global warming

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potential (GWP) than carbon dioxide (CO_2) over a 100-year timespan. N_2O alone contributed 5% to the total anthropogenic GHG emissions in 2010 (IPCC 2014). In addition, N_2O is the single most important ozone-depleting emission and is expected to remain the largest throughout the twenty-first century (Ravishankara et al. 2009). Global anthropogenic N_2O emissions are rapidly increasing with average annual emissions of 52 gigatons (Gt) CO_2 equivalent in 2011 and are expected to almost double by 2050 unless mitigation action is accelerated (UNEP 2013; IPCC 2014). The atmospheric concentration of N_2O exceeded the pre-industrial value by 20% and reached 324 parts per billion (ppb) in 2011. Nitric oxide (NO), on the other hand, is an environmental pollutant that participates in photochemical reactions in the troposphere that produce ozone and contribute to acid deposition (Davidson et al. 2000).

Agriculture is one of the major sources of anthropogenic N_2O emissions. Two-thirds of current anthropogenic N_2O emissions originate from agriculture (Smith et al. 2007; UNEP 2013). Agricultural emissions of both N_2O and NO are mainly associated with the use of nitrogen (N) fertilizers. They are produced in soils during the biochemical processes of nitrification and denitrification (Firestone and Davidson 1989; Davidson et al. 2000). N_2O emissions from agricultural soils show large variations due to differences in the environment, crops, and management (Lesschen et al. 2011). Moreover, the differences in measurement methodology are also responsible for the large variations in reported emissions. For example, extrapolation of the results of the discrete measurements done at weekly or biweekly intervals may either overestimate or underestimate total emissions compared to the results of continuous measurements. Fluxes estimated from continuous measurements are more reliable because they include all the temporal variations. Yet, the discrete measurement system is in common use and very few studies (Scheer et al. 2012; Weller et al. 2015) measured N_2O emissions using continuous measurement systems. Despite the large variations in emissions, N_2O direct soil emissions from agriculture are often estimated using the default Intergovernmental Panel on Climate Change (IPCC) emission factor (EF) of 1% of applied N (IPCC 2006). More intensive measurements are needed to get season-, site-, and crop-specific emissions factors.

More than 50% of applied N is not utilized by crops, posing huge economical costs and environmental concerns (Savant and Stangel 1990; Rochette et al. 2013). Therefore, N management research has focused on increasing use efficiency and crop productivity and reducing N_2O and NO emissions. Enhanced-efficiency fertilizers, such as those containing nitrification inhibitors and urease inhibitors, and slow-release fertilizers have been developed to increase the efficiency of fertilizer use by crops. They are also effective for mitigating N_2O and NO emissions (Akiyama et al. 2010). Similarly, enhanced-efficiency fertilizer application methods, such as urea deep placement (UDP), reduce N losses up to 35% and increase rice yield up to 20% compared to conventional broadcast applications (Savant and Stangel 1990; Huda et al. 2016; Miah et al. 2016). UDP increases nitrogen use efficiency (NUE) because it minimizes N losses from surface runoff and ammonia volatilization as the ammonium is protected from nitrification/denitrification in anaerobic soil layers. However, studies on the effects of UDP on N_2O and NO emissions are still lacking, particularly in flooded rice fields. In our previous study (Gaihre et al. 2015), UDP reduced N_2O emissions by up to 84% compared to broadcast applications during the dry season. In the recent meta-analysis, van Kessel et al. (2013) found that placement of fertilizer-N at > 5 cm depth reduced N_2O emissions in humid climates under no-tillage or reduced tillage conditions. Contrary to these results, Linquist et al. (2012) found that deep placement or banding of fertilizer N in continuously flooded rice systems increased N_2O emissions by 18%.

Limited field studies report variable results on the effect of UDP on N_2O and NO emissions. Therefore, the evidence is still inconclusive to predict the effects of UDP on N_2O and NO emissions in rice fields. More field studies under different water management regimes are needed to come up with comprehensive conclusions. In this paper, we present fluxes of N_2O and NO measured continuously throughout rice-growing and fallow seasons using automated closed chamber techniques from intensive rice cropping systems in Bangladesh with the following specific objectives:

- Assess the seasonal and spatial variations in N_2O and NO emissions under different N-fertilizer placement methods.

- Compare the N_2O and NO emissions and NUE from UDP versus broadcast urea.
- Determine the effects of AWD irrigation on grain yields and NUE under different N fertilizer applications.

Materials and methods

Experimental site and weather conditions

The field experiments were conducted at two locations in Bangladesh—Bangladesh Agricultural University (BAU), Mymensingh (latitude: 24°42′55″; longitude: 90°25′47″), and Bangladesh Rice Research Institute (BRRI), Gazipur (latitude: 23°59′25″, longitude: 90°24′33″)—during two consecutive rice-growing seasons (*Aus* and *Aman*) in 2014. The study sites were under triple rice cropping systems. *Aus* (pre-monsoon, May–August) and *Aman* (monsoon, August–November) are considered wet seasons (rainfed rice), where monsoon rain is typically sufficient for rice production. The climate is humid sub-tropical monsoon. Average annual rainfall is ca. 1500 mm, primarily received from June to October. Daily rainfall and air temperature for two locations during two growing seasons are shown in Fig. 1. The soil at BAU had relatively high organic carbon (1.67 vs. 1.27% at BRRI) and low phosphorus content (2.54 vs. 11.47 mg kg^{-1}) compared to the BRRI soil (Table 1).

Experimental design and treatments

Automated continuous N_2O and NO measurements were conducted at both sites with triple rice cropping systems. At each site, N-fertilizer treatments were arranged in a randomized split-plot design with three replications. Water regimes, i.e., continuous standing water (CSW) and AWD were considered as main plots, while fertilizer treatments were as sub-plots. Experiments were established in 2013 (Gaihre et al. 2015) and the treatments' randomization kept constant for each season. The N-fertilizer treatments included: (1) control: no N; (2) urea briquette (UB) application: deep placement of 52 kg N ha^{-1}; and (3) prilled urea (PU) application: broadcast application of 78 kg N ha^{-1} during the *Aus* season and 52 kg N ha^{-1} during the *Aman* season. Each experimental plot was 5.6 m × 3.6 m at BAU and 4.3 m × 3.2 m at BRRI.

UDP and broadcast applications of PU (also includes granular urea) in two to three splits are commonly used N-fertilizer application methods in Bangladesh. Urea briquettes of 1.8 g for *Aus* and *Aman* seasons are deep-placed 7–10 cm in depth at 40 cm × 40 cm spacing between four hills of rice at every alternate row. This results in 62,500 placement sites per ha and supplies 52 kg N ha^{-1} for wet seasons (*Aus* and *Aman*). UDP saves 30–35% urea compared with surface broadcast application and increases grain yield by up to 20%; therefore, the N rate for UDP is 30% less compared with broadcast PU (at the recommended dose) (Savant and Stangel 1990; Kapoor et al.

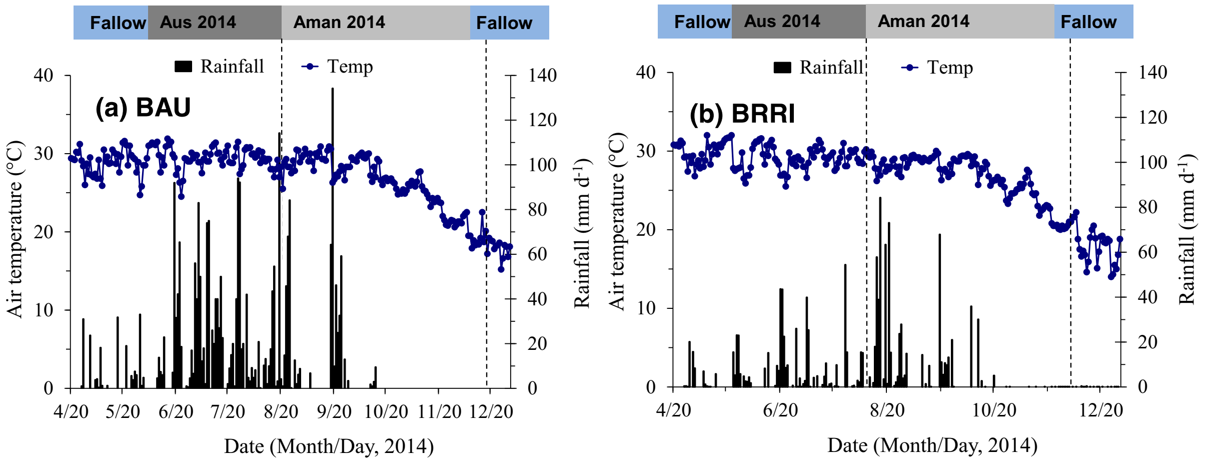

Fig. 1 Daily average of rainfall and air temperature during *Aus* and *Aman* rice-growing seasons at **a** Bangladesh Agricultural University (BAU) and **b** Bangladesh Rice Research Institute (BRRI)

Table 1 Physicochemical properties of soil used in the experiments

Soil property	BAU	BRRI
pH-H_2O	5.7	5.8
Organic carbon (%)	1.67	1.27
Total N (%)	0.16	0.14
Available P (mg/kg)	2.54	11.47
Available K ($cmol_c\ kg^{-1}$)	0.10	0.12
Available S (mg/kg)	12	–
Particle size (%)		
Sand	11.44	29.96
Silt	72.4	40.10
Clay	16.16	29.94

2008; FRG 2012; Gregory et al. 2010). Deep placement was done as a single application during the first topdressing (7–10 days after transplanting) of PU. PU was applied in two splits during the *Aus* season and three splits (BAU) during the *Aman* season.

Agronomic practice

The cropping period—from planting to harvest—was from May to August for the *Aus* season and from August to December for the *Aman* season. In both seasons, phosphorus (P; triple superphosphate) and potassium (K; muriate of potash) fertilizers were applied basally in all the plots during final land preparation at 16 kg P ha^{-1} and 42 kg K ha^{-1}. In addition, at BAU sulfur (S) and zinc (Zn) were applied to all plots at a rate of 20 kg S ha^{-1} as gypsum and 3 kg Zn ha^{-1} as zinc oxide but not at BRRI. At each site and season, 3- to 4-week-old rice seedlings were manually transplanted into the puddled soils with 20 cm × 20 cm distance between hills. Planted rice varieties were BRRI dhan 48 and BR 22 for *Aus* and *Aman* seasons, respectively, at both sites. Paddy rice of CSW plots were kept continuously flooded until 2 weeks before harvesting (Fig. 2). While plots under AWD conditions were irrigated following AWD principles, they were irrigated when the water depth in AWD pipes was 12–15 cm below soil surface. AWD irrigation was implemented 1 week after the first topdressing of PU or UDP and continued until 2 weeks before harvesting. But all plots were maintained continuously flooded for a week after the second and third topdressing during topdressing of PU and during the flowering stage. However, *Aus* and *Aman* are wet seasons, and drying of AWD plots was merely due to chance, depending on timing of rainfall.

Measurement of N_2O and NO emissions

Automated GHG sampling and analysis systems were continuously conducted from June 2013 to December 2014, covering five consecutive rice-growing seasons. A detailed description of the measuring system was reported earlier (Fig. 3; Gaihre et al. 2015). In brief, an automated GHG measurement system consisted of 12 plexiglass chambers. Each chamber was installed in a plot between two rows of rice over a fixed aluminum base that covered a surface area of 0.148 m^2 and headspace volume of 0.0578 m^3 (57.8 L). Since rice plants were not covered by the chamber, the height of the chamber was kept same throughout the growing season. However, chamber saturation or dilution effects were not observed. Chambers were connected to N_2O and NO analyzers for on-site measurements. Out of 12 chambers, nine chambers were installed under CSW plots (three treatments, three replications), and three chambers were installed under AWD plots. Air samples from each chamber were taken six times at 8-min intervals (0, 8, 16, 24, 32, and 40 min), which resulted in a chamber closing time of 40 min. Each chamber was sampled every 3 h, which allowed calculation of eight fluxes per day. There were three sets of four chambers, one set (one replication), i.e., four chambers were sampled in a 1-h cycle. The 3-h sequence consisted of three 1-h sequences that were identical, except the first hour applied to Chambers 1–4 (Rep 1), the second hour applied to Chambers 5–8 (Rep 2), and the third hour applied to Chambers 9–12 (Rep 3). Chambers 4, 8, and 12 represent the non-replicated treatments of the AWD experiment. N_2O and NO emissions measurement from AWD experiment are not reported here in.

Air samples collected from the chambers were passed to the analyzers using a Teflon tube with ¼″ diameter via a 13-port sample manifold (equipped with solenoid valve, i.e., KIP valves). Out of 13 valves in a sample manifold, 12 were used for air samples of the respective 12 chambers, while the last one was used for the calibration gases (i.e., connected to

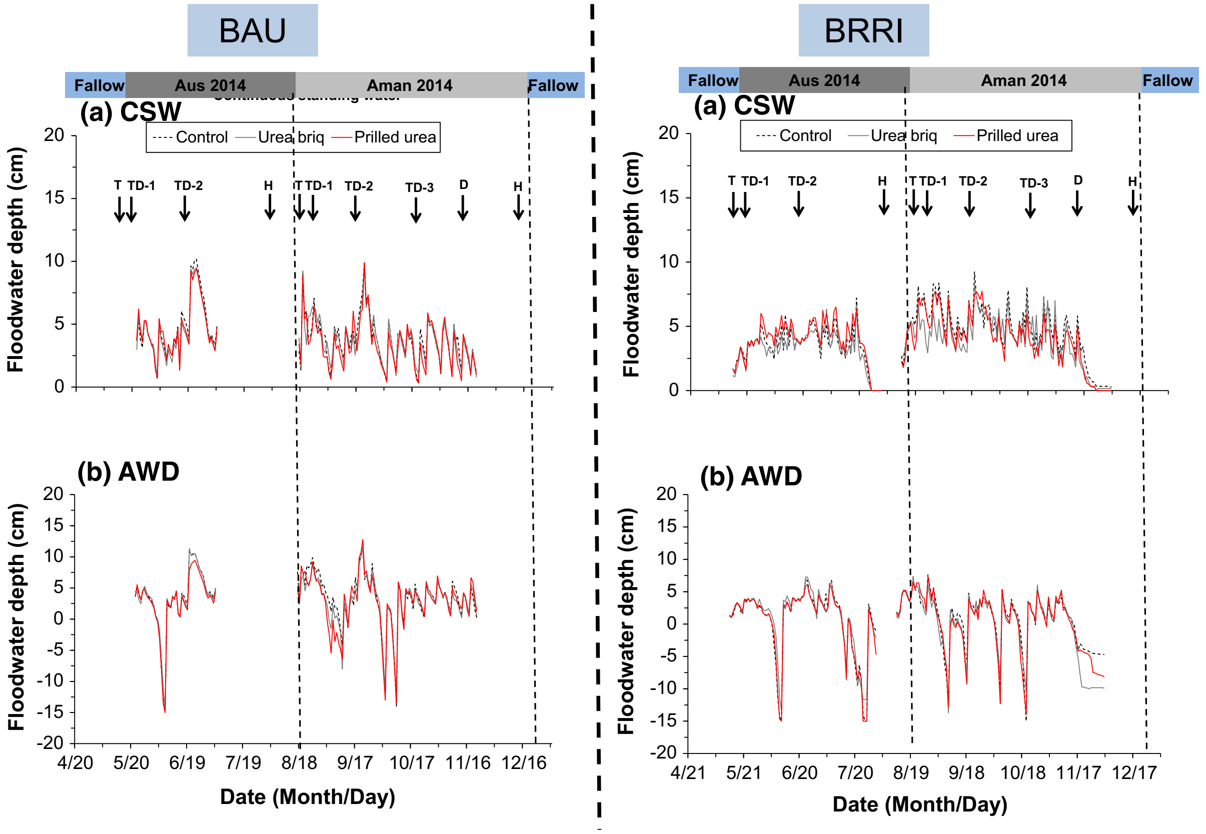

Fig. 2 Floodwater depth during *Aus* and *Aman* rice-growing seasons at **a** Bangladesh Agricultural University (BAU) and **b** Bangladesh Rice Research Institute (BRRI). T, TD-1, TD-2, TD-3, D, and H are transplanting, first, second, and third topdressing, final drainage, and harvesting, respectively

calibrator). Each sample valve in the manifold was controlled by a datalogger (Campbell Scientific, CR3000) via 16 port (channel) relay controller (SDMCD16 AC/DC Relay Controller). Air samples were filtered and dried before passing to the respective analyzers.

N_2O concentration of the air sample was measured by a Teledyne Advanced Pollution Instrumentation (API) T320U Gas Filter Correlation Analyzer. The T320U uses an infrared radiation (IR) absorption principle. IR travels through a sample chamber in the analyzer filled with gas bearing a varying concentration of N_2O (air sample or calibration gases). The sample and calibration gases should be supplied at ambient atmospheric pressure to establish a stable gas flow through the sample chamber where gases ability to absorb infrared radiation is measured. It can analyze N_2O concentration up to 200 ppm with lower detection limit of $<$ 10 ppb. NO concentration of the air sample was measured by a Teledyne API T200 Nitrogen Oxide Analyzer. The T200 uses a chemilumінescent detection principle. It measures the amount of NO present in a gas by detecting the chemiluminescence that occurs when nitrogen oxide is exposed to ozone. It can analyze NO concentration up to 20 ppm with lower detection limit of 0.4 ppb.

Both N_2O and NO analyzers were calibrated weekly using a Teledyne T700 Dynamic Dilution Calibrator. The N_2O analyzer was calibrated for two ranges of concentration, i.e., low range (1600 ppb) and high range (8000 ppb). Similarly, NO analyzer was also calibrated for two ranges of concentration, i.e., low range (40 ppb) and high range (400 ppb). For both N_2O and NO concentration, the high range values were used when concentration of air sample exceeded the low range. The minimum detection limits of the flux measured based on the chambers used (57.8 L volume, 0.146 m^2 area, and 40 min chamber closure

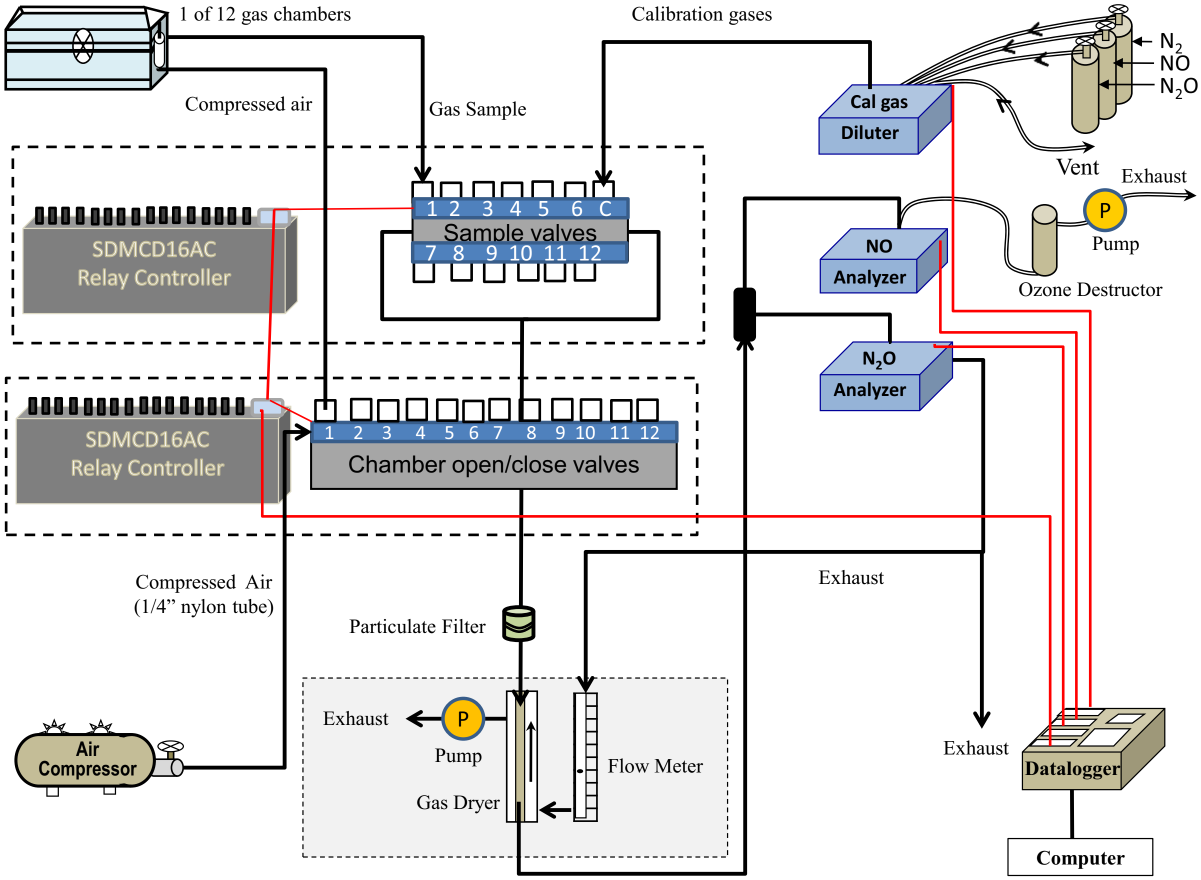

Fig. 3 Schematic diagram showing different components of the automated continuous N_2O and NO measurement system

time) were 17 and 4 µg m^2 h^{-1} for N_2O and NO, respectively.

The N_2O and NO fluxes were calculated from the slope of the linear increase or decrease in concentration of respective gases against the chamber closure time. The slope (ppb min^{-1}) is then corrected for air temperature, atmospheric pressure, and the ratio of chamber volume to surface area. An emission event was considered significant when the slope was significant at $p < 0.05$. Therefore, emissions with non-significant slopes were discarded. Cumulative seasonal total emissions (g N_2O-N or NO-N) were calculated summing the hourly emission rates. Yield-scaled N_2O and NO emissions (g N t^{-1} grain) were calculated from the ratio of seasonal total emissions to grain yields. Direct N_2O/NO emission factors (EF_d %) of applied N fertilizer were calculated by using the following equation:

$$EFd\% \ = [EF - E0]100/N\ Rate$$

where EF (kg N ha^{-1}) is the cumulative N_2O or NO flux from the N-fertilized treatment, and E0 (kg N ha^{-1}) is the cumulative N_2O emission from the control treatment.

Rice plants at physiological maturity were harvested from each plot (5 m^2 area) and recorded grain yields (14% moisture content). Grain and straw components were separated and dried at 70 °C to a constant weight. Both grain and straw samples were analyzed for their N content and the total N uptake was calculated.

Data analysis

Analysis of variance (ANOVA) for the different response variables was performed with SAS 9.3, using Generalized Linear Mixed Models. The ANOVA model had a split plot structure where the main plot was represented by the site and the subplot was represented by the treatment in the case of response variables associated with N_2O or NO emissions or by combinations of the treatment and water regime in the case of grain yield and nitrogen efficiency. Site, treatment, water regime, and their interactions were handled as fixed effects, while replications and the error terms rep × site and rep × site × treatment or rep × site × treatment × water regime were handled as random effects. ANOVA analysis for emission factors (EF_d %), agronomic nitrogen efficiency (AE_N), and nitrogen recovery efficiency (RE_N), which are calculated only for the PU and UDP treatments, was conducted with the residual as the error term to obtain enough denominator error degrees of freedom for an appropriate significance test. ANOVA involving model comparisons also used a split plot structured with the site as the main plot as explained above. Pre-planned comparisons (contrasts) of the linear and exponential estimates of N_2O emissions under each of the three treatments and at the two locations were carried out independently of the ANOVA results, based on literature reports indicating N_2O emission underestimations by the linear model and based on analysis done previously with data from the same experiment collected during the *Boro* 2014 season. A pairwise comparison of means from significant effects in the ANOVA was conducted with an LSD test.

Our previous study and literature (Venterea et al. 2009) showed that the exponential model estimated consistently higher emissions than linear estimates. Therefore, in addition to linear emission estimates, fluxes for N_2O were also estimated with the exponential model:

$$y = i + a\, e^{-bt}; \quad I = i + a$$

where y is the N_2O concentration inside the chamber in ppb, i and a are adjusting parameters, I or actual intercept estimates the background N_2O concentration at time 0 ($I = i + a$), b is a parameter associated with the rate of concentration change per minute, and t is the time 0–40 min at 8-min intervals.

ANOVA was conducted to test the fixed effects of treatments, site, and model, and their interaction on emissions. Based on the hypothesis that the exponential model estimates higher emissions than the linear model, planned comparisons were performed between two models within treatment and site.

Results

Seasonal variations of N_2O fluxes

N_2O fluxes were discrete and sporadic throughout the rice-growing and fallow seasons. They were event-specific, i.e., observed only after topdressing of PU, during the drying period, and after reflooding of the dry soil for land preparation for the next crop. Except during peak emission events, N_2O fluxes were within a range of ± 40 μg N m^{-2} h^{-1}.

At BAU, peaks in N_2O fluxes were observed in broadcast PU treatments after 2–5 days of topdressing and continued from a few days to a week. However, patterns and magnitudes of emission peaks were not consistent across fertilizer applications and seasons. The magnitudes of fertilizer-induced peaks were higher during the *Aus* season, while they were very small to negligible during the *Aman* season (Fig. 4a). The highest fertilizer-induced emission peak (288 μg N m^{-2} h^{-1}) was observed after the second

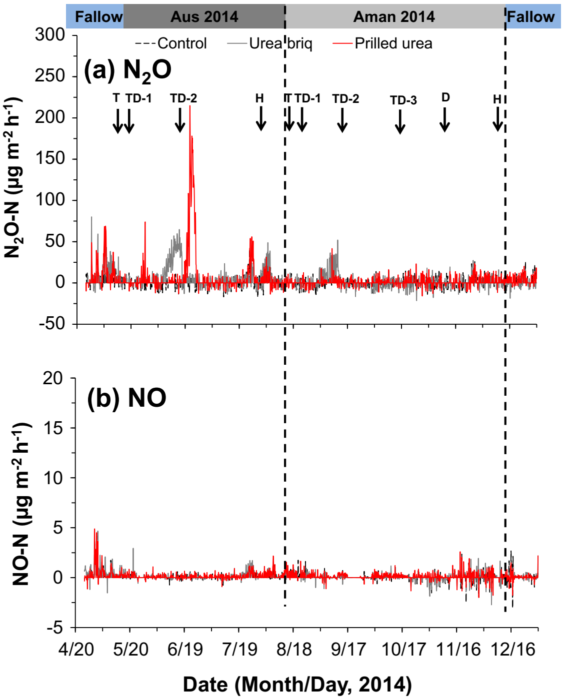

Fig. 4 Seasonal variations in N_2O and NO emission rates under different fertilizer treatments during *Aus*, *Aman*, and fallow seasons, 2014 at BAU. T, TD-1, TD-2, TD-3, D, and H are transplanting, first, second, and third topdressing, final drainage, and harvesting, respectively

topdressing of PU in the *Aus* season. On the other hand, fertilizer-induced emission peaks in UDP treatments were observed for a week (June 7–16) during the *Aus* season after 3 weeks of UDP but not during the *Aman* season. However, it should be noted that this was mainly caused by exceptionally high emissions from one single chamber after UDP on May 22. The rest of the time, patterns and magnitudes of fluxes were similar between control and UDP treatments. Over the rice-growing season (irrespective of the treatments), N_2O fluxes ranged from − 17 to 215 μg N m^{-2} h^{-1} during the *Aus* season, while they ranged from − 29 to 55 μg N m^{-2} h^{-1} during the *Aman* season.

At BRRI, fertilizer-induced emission peaks in broadcast PU treatments were prominent only after the second topdressing during the *Aus* season. As in BAU, emission peaks after the first and second topdressing during the *Aman* season were negligible. Similarly, some emission peaks were observed in UDP treatments during the *Aus* season but not during the *Aman* season (Fig. 5a). Overall, N_2O fluxes ranged from − 15 to 99 μg N m^{-2} h^{-1} during the *Aus* season, while they ranged from − 21 to 63 μg N m^{-2} h^{-1} during the *Aman* season. Exceptionally higher emission peaks (1247–1865 μg N m^{-2} h^{-1}) were observed at fallow (land preparation for *Aus* transplanting). Some emission peaks occurred after final drainage for harvest of *Aman* rice.

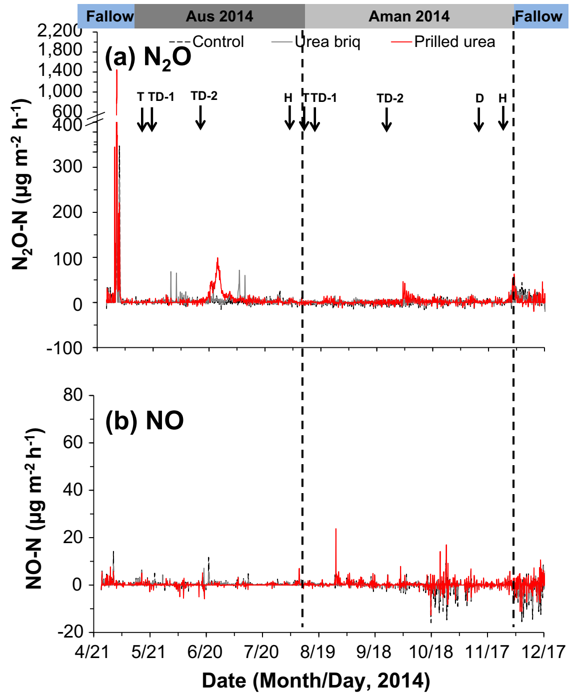

Fig. 5 Seasonal variations in N_2O and NO emission rates under different fertilizer treatments during *Aus*, *Aman*, and fallow seasons, 2014 at BRRI. T, TD-1, TD-2, TD-3, D, and H are transplanting, first, second, and third topdressing, final drainage, and harvesting, respectively

Seasonal variations of NO fluxes

Seasonal variations of NO fluxes from all the treatments during two rice-growing seasons (*Aus*–*Aman* 2014), including fallow seasons at BAU and BRRI sites, are displayed in Figs. 4b and 5b. At both sites, effects of N fertilizer on emissions were not apparent during the *Aus* or *Aman* seasons. Negative fluxes were observed, particularly at night during October–December, with wider variations at BRRI compared to BAU. Fluxes were relatively higher at BRRI compared with BAU. NO fluxes ranged from − 2 to 3 μg N m^{-2} h^{-1} at BAU and from − 31 to 40 μg N m^{-2} h^{-1} at BRRI. Emissions patterns showed seasonal differences; they were relatively higher during the *Aman* season than the *Aus* season (Fig. 5b).

Cumulative N_2O and NO emissions

Cumulative N_2O emissions, irrespective of sites, ranged from 15 to 174 g N ha^{-1} during the *Aus* season and from 9 to 81 g N ha^{-1} during the *Aman* season (excluding fallow season emissions) (Table 2). Emissions varied significantly among fertilizer treatments and sites. Effects of N fertilizer on emissions were season-specific; it increased emissions during the *Aus* season ($p < 0.09$) but not during the *Aman* season. However, emissions between UB and broadcast PU treatments were similar. In the *Aman* season, average emissions across treatments at BRRI were higher than at BAU. Fertilizer-induced EF ranged from 0.10 to 0.20% during the *Aus* season and from 0.02 to 0.06% during the *Aman* season. As with absolute emissions, yield-scaled emissions (g N_2O-N t^{-1} grain) were affected by N fertilizer treatments during the *Aus* season and by site during the *Aman* season. The combined analysis of N_2O emissions across two sites showed no significant interaction effects of fertilizer

Table 2 Seasonal N_2O and NO emissions at all N-fertilizer treatments under continuous standing water (CSW) regime at BAU and BRRI

Site	N placement	N_2O emissions			EF-N N_2O-N %, w/w	NO emissions		EF-NO-N %, w/w
		g N ha^{-1}	kg CO_2 eq. ha^{-1}	g N t^{-1} grain		g N ha^{-1}	g N t^{-1} grain	
Aus 2014								
BAU	Control	15.71	6.37	3.79	–	1.01a	0.27a	–
	UB	67.40	28.06	15.70	0.100	2.04a	0.47a	0.002
	PU	173.81	72.38	42.44	0.203	2.33a	0.56a	0.002
BRRI	Control	24.54	11.49	6.42	–	7.91a	2.06a	–
	UB	79.94	37.44	17.40	0.106	6.86ab	1.52b	− 0.002
	PU	136.97	64.14	29.02	0.144	3.00c	0.65c	− 0.006
All	Control	19.92b	8.93b	5.10b				
	UB	73.67ab	32.75ab	16.55ab				
	PU	155.39a	68.26a	35.73a				
BAU	All							0.0018a
BRRI								− 0.0042b
ANOVA (p values)								
Site (S)		0.9118	0.9199	0.7600	0.8028	0.1078	0.0960	0.0141
Treatment (T)		0.0855	0.1008	0.0733	0.5106	0.0654	0.0533	0.2682
S × T		0.8795	0.9338	0.7539	0.7568	0.0084	0.0073	0.3306
Aman 2014								
	Control	28.53	13.12	8.20	–	− 5.12	− 1.51	–
All	UB	50.32	22.97	11.83	0.0419	− 0.61	− 0.19	0.0087
	PU	60.93	27.48	14.46	0.0623	6.38	1.51	0.0221
BAU	All	24.27b	10.11b	5.47b				
BRRI		68.92a	32.27a	17.52a				
ANOVA (p values)								
Site (S)		0.0252	0.0184	0.0199	0.7816	0.7838	0.7707	0.2461
Treatment (T)		0.3247	0.3531	0.5219	0.7476	0.1880	0.2486	0.4531
S × T		0.9179	0.9113	0.9435	0.8350	0.3942	0.4230	0.5750

Within a column and season for each response variable, means followed by same letters are not significantly different at 5% level of probablity. UB and PU represent deep placement of urea briquettes and broadcast prilled urea, respectively

treatment and site on any of the response variables (Table 2).

Cumulative and yield-scaled NO emissions were very low, which varied within the range of ± 10 g N ha^{-1} and ± 3 g N t^{-1} grain yield, respectively. In the *Aus* season, emissions were significantly affected by fertilizer treatments ($p < 0.06$–0.07) and fertilizer × site interactions ($p < 0.01$). Both cumulative emissions and yield-scaled emissions were higher from the control than from fertilized treatments at BRRI. But emissions during the *Aman* season were not affected by N-fertilizer treatments, sites, or their interactions.

Linear and exponential model estimates of cumulative N_2O emissions

Exponential model estimates of fluxes followed similar patterns with the linear regression model. The treatment difference between PU and UDP was not detected by any of the flux calculation schemes. Though the magnitude of total N_2O emissions differences predicted by the linear and exponential regression models were small, emissions calculated by exponential models were significantly higher than that of emissions calculated by linear models (Tables 3, 4). Planned comparison showed that the exponential model estimated higher N_2O emissions by 11 g ha^{-1} during the *Aman* 2014 season at both locations.

Grain yield and nitrogen use efficiency

N-fertilizer application significantly affected grain yields. Without N fertilizer, grain yields ranged from 3.09 to 3.86 t ha^{-1}. In the *Aus* season, across water regimes and sites, UDP and broadcast PU increased grain yields by 22 and 18%, respectively (Table 5). However, the yield difference (4%) between UDP and PU was not significantly different. Grain yield at BRRI (4.22 t ha^{-1}) was significantly higher ($p < 0.01$) than BAU (3.85 t ha^{-1}). Interactions of fertilizer treatment with either site or water were not significant. On the other hand, in the *Aman* season, there were significant interaction effects of N fertilizer with site on grain yields (Table 4). UDP increased grain yields significantly compared to PU at BAU but not at BRRI. Across water regimes, UDP and broadcast PU increased grain yields over the control by 56 and 38% at BAU and by 16 and 14% at BRRI, respectively. Water regime interacted with the site; AWD irrigation significantly reduced grain yields at BRRI but not at BAU.

Across water regimes and sites, deep placement significantly increased agronomic use (AE_N) efficiency (15 kg grain kg^{-1} N applied) compared to PU (8 kg grain kg^{-1} N applied) during the *Aus* season. On the other hand, N fertilizer and water regime had significant interaction with site during the *Aman* season. Both UDP and AWD increased AE_N at BAU but not at BRRI (Table 5). Likewise, UDP increased RE_N during both the *Aus* and *Aman* seasons at BAU. On the other hand, AWD irrigation during the *Aman* season significantly ($p < 0.05$) reduced the RE_N at BRRI, while they were similar between CSW and AWD at BAU.

Table 3 Test of fixed effects of site, treatment, model, and their interaction on cumulative N_2O emissions during the *Aman* 2014 season at BAU and BRRI

Effect	Num DF	Den DF	F value	Pr > F
Site	1	4.75	0.00	0.9494
Treatment (T)	2	5.35	1.61	0.2844
Site × T	2	5.76	0.13	0.8765
Model	1	12	11.88	0.0048
Model × T	2	12	0.86	0.4491
Site × model	1	12	0.01	0.9258
Site × model × T	2	12	1.99	0.1796

Discussion

N_2O and NO emissions

UDP is recommended for lowland rice cultivation to reduce N losses to the environment and increase NUE. Placement of urea briquettes at 7–10 cm depth concentrates NH_4^+-N in the anaerobic soil layer and thus reduces its movement to the soil surface or in floodwater (Savant and Stangel 1990; Kapoor et al. 2008; Rochette et al. 2013; Huda et al. 2016). This results in reduction of potential nitrification/denitrification losses and thus N_2O and NO emissions. This hypothesis is supported by previous studies that N_2O is mainly produced near the soil surface compared with a deeper soil layer (Yoh et al. 1997; Venterea and Stanenas 2008). Contrary to UDP, broadcast application of PU produces some N_2O emission peaks after 2–5 days of its application (Figs. 4a, 5a), because the NH_4^+-N is subjected to nitrification and denitrification even under flooded (anaerobic) conditions. The fertilizer-induced N_2O emission peaks observed in this study in the broadcast PU treatment are consistent with previous studies (Bronson et al. 1997; Chen et al. 1997; Sander et al. 2014; Weller et al. 2016). On the other hand, the fertilizer-induced emission peaks were rarely observed in UDP plots at both sites. The small but delayed emission peaks that occurred in the *Aus* season could be due to improper placement/closing of the urea briquettes with soil or disturbances caused during weeding, which might have released some NH_4^+-N to the soil surface.

N_2O emissions showed higher seasonal and spatial variations, which are consistent with previous studies (Akiyama et al. 2005; Scheer et al. 2012; Liang et al. 2013). The emission peaks after topdressing of PU were not consistent across sites, or seasons (Figs. 4, 5). They were higher at BAU site compared with BRRI site, particularly during the *Aus* season. While

Table 4 Planned comparison of seasonal cumulative emissions between linear and exponential estimates within treatment and site

Site	N placement	Model		Difference (g N ha^{-1})	Standard error	DF	t value	Pr > t
Both	Control	Exp	Lin	17.50	5.83	12	3.0	0.0112
	UB	Exp	Lin	6.85	5.83	12	1.17	0.2633
	PU	Exp	Lin	10.52	5.83	12	1.80	0.0967
BAU	All	Exp	Lin	11.94	4.76	12	2.50	0.0277
BRRI	All	Exp	Lin	11.29	4.76	12	2.37	0.0354

emission peaks were smaller to negligible at both sites during *Aman* season. Despite the N_2O emission peaks after broadcast PU, during dry or drained periods, the contributions of fertilizer-induced N_2O emissions to seasonal total emissions were not sufficient to result in significant difference over UDP. Application of N fertilizer increased emissions by up to 11 times compared to the emissions from the control (without N fertilizer), particularly during the *Aus* season. However, emissions from PU and UB treatments were not significantly different across seasons and sites. Some previous studies have also shown no difference in N_2O emissions between UDP and broadcast application, particularly under optimum N rates (Adviento-Borbe and Linquist 2016).

Lower N_2O emission factors (< 0.2) observed in this study for broadcast PU are in agreement with previous studies in continuously flooded rice systems (Bronson et al. 1997; Chen et al. 1997). In the recent meta-analysis, Linquist et al. (2012) found that N fertilizer-induced N_2O emission factors during the rice-growing season were 0.21% for continuously flooded fields and 0.40% for fields with intermittent drainage. Likewise, in a review, Akiyama et al. (2005) reported emission factors of 0.22 and 0.37% for rice fields with continuous flooding and mid-season drainage, respectively. In continuously flooded soils, denitrification provides more opportunity for the reduction of N_2O to N_2 (Firestone and Davidson 1989). However, it is reported that emissions increase when N is applied in excess, i.e., beyond plant uptake (Kim et al. 2013). Lower emission factors observed in this study may be attributed to N rates, other losses to the environment and weather condition. Nitrogen rates were optimum for both broadcast and deep placement. Moreover, in the wet seasons, N is lost mainly as ammonia volatilization and surface runoff resulting in less N loss as N_2O and NO emissions. In this study site, ammonia volatilization loss accounted for up to 20% of applied N fertilizer (Huda et al. 2016). In our previous study, N_2O emissions in broadcast PU increased significantly in the dry season, whereas emissions were consistently lower in UB treatment irrespective of rice-growing seasons (Gaihre et al. 2015). Nevertheless, emissions during wet seasons were similar between UDP and broadcast urea. These results confirm that N_2O and NO emissions from continuously flooded rice fields during the wet season are negligible under current rates and methods of N applications. Since *Aus* and *Aman* rice in Bangladesh are wet season rice and remain continuously flooded most of the time during rice-growing seasons, their contribution to N_2O and NO emissions could be insignificant compared to a default emission factor of 1% of applied N.

Magnitudes and patterns of emissions may also vary with flux calculation schemes, i.e., use of linear or exponential models (Venterea et al. 2009). However, in this study, the prediction of differences in total N_2O emissions between the two regression models were not large in spite of the exponential model producing more significant events. This is explained by the increase in the number of negative emissions offset by a similar increase in the number of positive emissions with the exponential model as compared with the linear model. The capability of the exponential model to identify larger numbers of significant emission events can be interpreted as an advantage, relative to the linear model, with the potential of providing opportunities to explain the emission events and to identify the factors involved in N_2O emissions. The large number of significant events with negative slopes from the exponential model can be useful to study the mechanisms in place for the soil to act as a N_2O sink. Fewer

Table 5 Grain yields, agronomic nitrogen use efficiency (AE$_N$) and recovery efficiency (RE$_N$) at all N fertilizer treatments under continuous standing water (CSW) and alternate wetting and drying (AWD) water regimes at BAU and BRRI

Site	Water regimes	N placement	Grain yield (t ha^{-1})	AE$_N$ (kg grain kg N^{-1}	RE$_N$ (kg N uptake kg N^{-1}
Aus 2014					
BAU	All	Control	3.33	–	–
		UB	4.23	17.46	0.61a
		PU	3.99	8.49	0.25b
BRRI	All	Control	3.79	–	–
		UB	4.44	12.46	0.38a
		PU	4.43	8.20	0.38a
All	All	Control	3.56b	–	
		UB	4.34a	14.96a	
		PU	4.21a	8.34b	
ANOVA (p values)					
Site (S)			0.0008	0.4145	0.5291
Water (W)			0.0683	0.2475	0.2205
Treatment (T)			< 0.0001	0.0239	0.0291
T × W			0.6422	0.6734	0.3941
T × S			0.4919	0.3767	0.0270
S × W			0.5604	0.3202	0.5656
T × W × S			0.7281	0.8274	0.6773
Aman 2014					
BAU	All	Control	3.31c	–	–
		UB	5.15a	35.37a	0.73a
		PU	4.58b	24.46b	0.16b
BRRI	All	Control	3.33b	–	–
		UB	3.85a	10.03a	0.30a
		PU	3.81a	9.17a	0.22a
BAU	CSW	All	4.24a	24.24b	0.41a
	AWD		4.46a	35.28a	0.48a
BRRI	CSW	All	3.93a	11.23a	0.32a
	AWD		3.41b	7.97a	0.20b
ANOVA (p values)					
Site (S)			0.0030	< 0.0001	0.0094
Water (W)			0.0783	0.0701	0.5398
Treatment (T)			< 0.0001	0.0075	< 0.0001
T × W			0.4380	0.5683	0.3990
T × S			< 0.0001	0.0189	< 0.0001
S × W			0.0001	0.0022	0.0373
T × W × S			0.1170	0.8608	0.4408

Within a column and season for each response variable, means followed by same letters are not significantly different at 5% level of probablity. UB and PU represent deep placement of urea briquettes and broadcast prilled urea, respectively

negative emissions with PU indicate more N was available for N_2O formation and emissions than with UDP or control treatments. Nevertheless, the exponential model estimates were consistently higher than the linear estimates.

NO emissions were very low to negligible irrespective of treatments, sites, and seasons (Table 2).

Occasional emissions observed after broadcast application of PU and during the dry period did not have significant contributions on cumulative emissions. For rice fields, there are very few studies that measured NO emissions. Our results are in agreement with Zhou et al. (2010); they observed very small amounts of NO emissions when the field was flooded during the rice-growing season compared with emissions during the wheat-growing season and fallow period. Significant NO fluxes were observed during mid-season drainage, which accounted for 85% of the total NO emissions. It is reported that NO is produced only at urea placement sites, and most of the produced NO was consumed instead of diffusing away from production sites in the soils (Hou et al. 2010). In flooded rice fields, NO could be reduced to N_2 during denitrification, resulting in lower cumulative emissions.

Grain yields and nitrogen use efficiency

The addition of N fertilizer significantly increased grain yields across sites and seasons, irrespective of application methods. However, the effects of placement methods on yields were site- and season-specific (Table 2). UDP increased grain yields over broadcast PU during the *Aman* season at the BAU site but not during the *Aus* season. The lack of yield response to N-fertilizer placement during the *Aus* season in this study is likely due the differences in N rates between UDP and broadcast PU. In the *Aus* season, N rate for UDP (52 kg ha^{-1}) was 33% less than that of broadcast PU (78 kg ha^{-1}), which is a common practice in Bangladesh. Contrary to these results, previous studies conducted across different seasons and sites in Bangladesh showed that 33% less N, as UDP compared to broadcast PU, increased grain yields by up to 20% (Gregory et al. 2010; Islam et al. 2016; Miah et al. 2016; Savant and Stangel 1990). In the *Aman* season, with the same N rates for broadcast PU and UDP (52 kg N ha^{-1}), UDP significantly increased grain yields compared to broadcast PU at the BAU site. But at the BRRI site, yields between UDP and broadcast PU were similar during both the *Aus* and *Aman* seasons. It is not clear why there is no yield response to application methods at BRRI. This site specific response could be associated with the weather condition (Fig. 1) and soil properties (Table 1). BRRI soil had lower organic C and higher amount of sand compared to BAU. It may be partially explained by increased N losses through leaching in both UDP and PU, because experimental plots drained water much faster (5.4 mm day^{-1} at BRRI vs. 3.0 mm day^{-1} at BAU) and were irrigated more frequently than at BAU (Fig. 2). Moreover, higher yields in control treatment (0 kg N ha^{-1}) suggest the higher residual N and/or indigenous N supply. Similar grain yields could be produced with lower N rate, i.e., < 52 kg ha^{-1}. Hence, deep placement of N did not show significant increase in yield over broadcast PU. We suggest further study at the BRRI site to determine whether lower yield response to additional N fertilizer was due to increased losses. Nevertheless, these results suggest that broadcast PU at 52 kg N ha^{-1} is not sufficient for optimum grain yields for the BAU site.

Higher yields in unfertilized plots (across water regimes) and the lower yield response to additional N fertilizer at BRRI resulted in lower NUE (AE_N and RE_N) compared to BAU. N recovery followed similar trend with grain yields. At BAU, UB increased RE_N from 25% (with broadcast PU) to 61% and from 25 to 73% for the *Aus* and *Aman* seasons, respectively. Similar results were observed by Huda et al. (2016) in which UB increased N recovery up to 64–67% from 35% with broadcast PU. Increased NUE with UB was also reported in West Africa (Bandaogo et al. 2014). As in grain yields, UB had no significant effects on RE_N at the BRRI site.

In this study, water regime had no significant effect on grain yields in any fertilizer treatment during the *Aus* season, because soil drying in AWD plots was very mild and did not affect rice growth and NUE. However, AWD significantly reduced grain yields and NUE during the *Aman* season at the BRRI site. Frequent drying of AWD plots might have created stress to rice plants. This resulted in significantly lower RE_N under AWD at BRRI compared to BAU. Contrary to these results, most of the previous studies have shown that the AWD irrigation practice has no significant effect on grain yield compared to farmer practice. In a recent review study, Lampayan et al. (2015) found that AWD saves irrigation water by up to 38% without significant effects on grain yields. Significant interaction of water regimes and sites suggests that the effects of AWD depend on frequency and intensity of drying and soil types. Moreover, safe AWD (in general, irrigate field when water level drops to about 15 cm below soil surface) should also be site specific to avoid stress to rice plant and yield penalty.

For example, BRRI site may need to irrigate before water level drops to about 12 cm below soil surface. Intense drying not only affects grain yields, but may have long-term impacts on soil fertility, particularly on soil organic carbon and N.

Conclusions

This study determined the effects of two nitrogen (N) placement methods in irrigated lowland rice systems on nitrous oxide (N_2O) and nitric oxide (NO) emissions, nitrogen use efficiency (NUE), and grain yields. Though broadcast prilled urea (PU) produced significant N_2O and NO emission peaks after topdressing, but there was no significant difference to total seasonal emissions between broadcast PU and urea deep placement (UDP). As with cumulative emissions, yield-scaled emissions and emission factors were similar between broadcast PU and UDP. NO emissions were very small compared with N_2O emissions during both rice-growing seasons. The effects of UDP on grain yields and NUE were site- and season-specific. UDP increased grain yields compared to broadcast PU at the BAU site during the *Aman* season. Similarly, UDP increased nitrogen recovery efficiency (RE_N) compared to broadcast PU at the BAU site during both the *Aus* and *Aman* seasons but not at the BRRI site. On the other hand, effects of alternate wetting and drying (AWD) irrigation was site and season specific. AWD had no effects on grain yields and RE_N during the *Aus* season at any site. But at BRRI, it decreased grain yield and RE_N in fertilized treatments during the *Aman* season.

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