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Impacts of urea deep placement on nitrous oxide and nitric oxide emissions from rice fields in Bangladesh

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ABSTRACT

Urea deep placement (UDP) increases nitrogen use efficiency (NUE) in lowland rice fields by reducing ammonia volatilization, surface runoff and increasing nitrogen uptake. However, its effects on N losses as nitrous oxide (N_2O) and nitric oxide (NO) are not yet clear. We conducted field experiments at two locations of Bangladesh — Bangladesh Agricultural University (BAU) and Bangladesh Rice Research Institute (BRRI) — to determine the effects of UDP vs broadcast urea on N_2O and NO emissions from rice fields.

N_2O and NO emissions were measured from three N fertilizer treatments (control [0 kg N/ha], UDP, broadcast urea) using automated gas sampling and analysis system continuously for three rice growing seasons — *Aus* (May–Aug), *Aman* (Aug–Dec) and *Boro* (Jan–May). Urea was applied as 2–3 split application, while for UDP treatment, urea briquettes were deep placed (7–10 cm depth) between 4 hills of rice at alternate rows to meet recommended N rates in a single application. Treatments were arranged in a randomized complete block design with three replications and N_2O and NO measurements were done at every three-hour interval.

N_2O emissions were sporadic and event specific. Peaks in N_2O emissions were observed after broadcast application of urea, during dry period and after re-flooding of the dry soil. For the rest of the time during the rice-growing season, emissions were very low to negligible. However, across the rice-growing seasons, UDP significantly ($P < 0.05$) reduced N_2O emissions compared with broadcast urea. Moreover, N_2O emissions showed significant spatial and seasonal variations. They were higher during *Boro* season compared with *Aus* and *Aman* seasons and at BAU site than that of BRRI. Conversely, emissions between *Aus* and *Aman* seasons and between control and UDP treatments were similar. In contrast to N_2O emissions, NO emissions were negligible and not affected by fertilizer treatment. However, significant spatial and seasonal variations were observed, with higher NO emissions at BRRI site compared with BAU and during *Boro* than that of *Aus* season.

1. Introduction

Nitrogen (N) plays a major role in crop production. Increases in N fertilizer use have boosted crop production to feed a growing world population. However, nitrogen use efficiency (NUE) in lowland rice fields is generally very low. The N loss is higher specifically when prilled urea (PU) is conventionally applied as broadcast where the recoveries of applied N are as low as 30–45% (Savant and Stangel, 1990; Sommer et al., 2004). More than 50% of applied N is not assimilated by plants and is lost through different mechanisms such as ammonia (NH_3) volatilization, surface runoff, leaching and nitrification–denitrification (Dong et al., 2012; Hayashi et al., 2006; Savant and Stangel, 1990; Singh et al., 1995; Watanabe et al., 2009; Zhao et al., 2009). Therefore, N use is associated with several negative environmental consequences, including increased emissions of nitrous oxide (N_2O) and nitric oxide (NO). N_2O, one of the major greenhouse gases responsible for global warming, is produced during nitrification and denitrification (Davidson et al., 2000; Firestone and Davidson, 1989). In addition to being a greenhouse gas, N_2O emission is the single most important ozone-depleting emission and is expected to remain the largest throughout the 21st century (Ravishankara et al., 2009). Moreover, nitrification–denitrification produces nitric oxide (NO), an environment pollutant that participates in photochemical reactions in the troposphere that produce ozone (Davidson et al., 2000).

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Atmospheric increase in N_2O concentration is mainly governed by anthropogenic sources. The agricultural sector is one of the major contributors, emitting about 60% of total anthropogenic N_2O (Smith et al., 2007). Emissions of N_2O and NO are mainly associated with N fertilizer use (or soil N content) and water regime. Generally, emissions increase with increasing N rates, particularly when N is applied in excess, i.e., beyond plant uptake. N_2O direct soil emissions from agriculture are often estimated using the default IPCC emission factor (EF) of 1% of applied N (IPCC, 2006). This EF is based on a large number of measurements (Bouwman et al., 2002; Stehfest and Bouwman, 2006) which lead to a mean value of 0.9%. Similarly, the global mean fertilizer-induced emission for NO is equivalent to 0.7% of applied N (Bouwman et al., 2002). Though IPCC (2006) considers the round value of 1% of applied N, N_2O emissions from agricultural soils show large temporal and spatial variations due to differences in environment, crops, and management (Lesschen et al., 2011). Moreover, the differences in measurement methodology also contribute to the large variations on reported emissions.

Generally, N_2O emissions are event-specific and appear only after irrigation (or rainfall, if upland crop), after N fertilization, and during the drying of flooded soil or during dry fallow periods (Bronson et al., 1997a, 1997b; Sander et al., 2014). The emission peaks appear only for a few hours to days. However, most of the studies measured emissions at weekly or biweekly intervals. The emissions reported from manual measurement might have missed the possible emission peaks if measurement frequency was not increased after fertilization and irrigation (Sander et al., 2014). Thus, extrapolation of the emissions (measured over wide intervals of time) over a season or a year may either over- or underestimate total emissions. On the other hand, automated continuous measurement includes all the temporal variations; it gives a real estimate of GHG fluxes, particularly for N_2O and NO. However, very limited studies (Scheer et al., 2012) measured N_2O emissions using automated continuous measurement systems. In this study, we report N_2O and NO emissions measured from rice fields using an automated continuous measurement system.

Lowland rice, which is cultivated in continuously flooded conditions, emits relatively less N_2O compared with upland crops (Akiyama et al., 2005, 2006). Because of continuous flooding of the soil, N_2O is further reduced to N_2 during denitrification (Davidson et al., 2000). However, due to increased application of N fertilizer and the change of irrigation practice from continuously flooded to water saving irrigation — alternate wetting and drying (AWD) rice cultivation may emit considerable amount of N_2O (Bronson et al., 1997a; Kim et al., 2013b). Despite the negative environmental consequences, N fertilizer is essential to increasing agricultural production to meet the food demands of a growing world population. Therefore, priority should be given to the best management practices, including optimum rate, source, timing and placement that increase crop productivity and NUE while reducing the negative environmental impacts such as water pollution, greenhouse effect, and ozone layer depletion.

N management studies have been conducted for many years, mainly with the aim of reducing N losses and increasing NUE. Improving the NUE and increasing agricultural productivity have been the major focus of research for the last one to two decades. However, the interest to reduce N_2O and NO emissions while increasing NUE is growing. Some of the N management practices are the use of slow- and controlled-release fertilizers, including polymer- and sulfur-coated fertilizers, nitrification inhibitors, urease inhibitors, and improved placement methods, which reduce the emissions of both gases, particularly N_2O from agricultural fields. Urea deep placement (UDP) is a promising technology that can drastically reduce N losses up to 35% and increase rice yield up to 20% (Mohanty et al., 1999; Savant and Stangel, 1990). UDP increases NUE by reducing N losses such as NH_3 volatilization (Rochette et al., 2013) and surface runoff and increasing plant uptake (Kapoor et al., 2008). UDP is gaining popularity for rice cultivation in some Asian countries such as Bangladesh (IFDC, 2012). Still, studies on the effects of UDP on N_2O and NO emissions are very limited. Reported studies have shown conflicting results. Therefore, this study was conducted to compare the N_2O and NO emissions from UDP vs. urea broadcast in intensive rice cropping systems and to assess their seasonal and spatial variations.

2. Materials and methods

2.1. Experimental site and weather conditions

The field experiments were conducted in two locations of 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 *Aus–Aman* 2013 and *Boro* 2014. The *Aus* (May–August) and *Aman* (August–November) seasons are considered as wet seasons (rainfed rice), where monsoon rain is typically sufficient for rice production. On the other hand, rice cultivation during the *Boro* (dry season, January–April) season is completely dependent on irrigation supply. The climate is humid sub-tropical monsoon. Average annual rainfall is ca. 1500 mm and primarily received from June to October. Daily rainfall and air temperature for the two locations during the three rice-growing seasons are shown in Fig. 1. The soil of BAU has relatively high organic C and low phosphorus content compared with BRRI soil. The physicochemical properties of the two soils before start of the experiments are shown in Table 1.

2.2. Experimental design and treatments

The three N fertilizer treatments were arranged in a randomized complete block design with three replications in each location. The randomization performed for the first season was the same for the two other seasons in both locations. Experimental plots were 5.6 m × 3.6 m at BAU and 4.3 m × 3.2 at BRRI. The N fertilizer treatments were as follows:

- Control: 0 kg N ha^{-1}
- Urea briquettes: deep placement of urea briquette at 52 kg N ha^{-1} during the *Aus* and *Aman* seasons and 78 kg N ha^{-1} during the *Boro* season.
- Prilled urea (PU): broadcast application at 78 kg N ha^{-1} during the *Aus* and *Aman* seasons or 104 kg N ha^{-1} in the *Boro* season.

Urea briquette (commonly called UDP) of 1.8 g (*Aus* and *Aman* seasons) and 2.7 g (*Boro* season) were deep placed (7– 10 cm depth) at 40 cm × 40 cm spacing (62, 500 placement sites per ha) between four hills of rice at every alternate row. It has been experimentally proven that UDP saves 30–35% urea compared with surface broadcast and produces yield increases of 20% or higher. Therefore, in this study, N rate for UDP is 30% less compared with broadcast PU (recommended dose) (FRG, 2012; Gregory et al., 2010; Kapoor et al., 2008; Savant and Stangel, 1990). Urea briquettes were deep placed as a single application during the first topdressing (7–17 days after transplanting, DAT) of PU. PU was applied as broadcast in two splits during the *Aus* and *Aman* seasons and three equal splits during the *Boro* season. The second and third topdressing of PU were done during maximum tillering (30–35 DAT) and panicle initiation (60–65 DAT) stages, respectively.

2.3. Crop management

Phosphorus (P) (triple superphosphate) and potassium (K) (muriate of potash) fertilizers were applied basally in all the plots during final land preparation at 16 and 42 kg ha^{-1} of P and K, respectively, in the *Aus–Aman* seasons. The same two nutrients were applied at 25 and 85 kg ha^{-1}, respectively, during the *Boro* season. In addition, sulfur

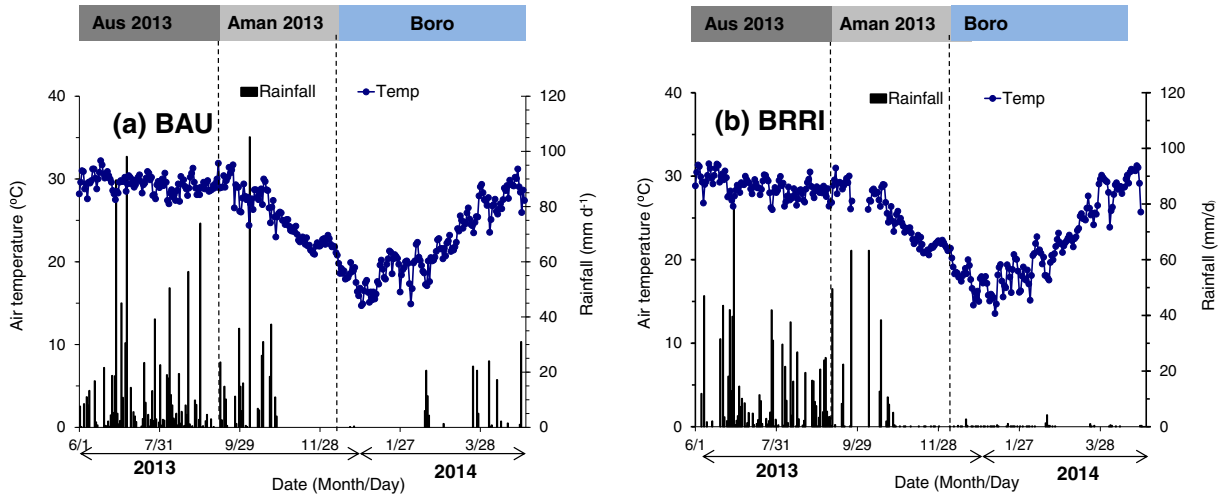

Fig. 1. Daily average of rainfall and air temperature during the *Aus*, *Aman* and *Boro* rice growing seasons at (a) Bangladesh Agricultural University (BAU) and (b) Bangladesh Rice Research Institute (BRRI).

(S) and zinc (Zn) were applied to all plots at the rate of 20 kg S ha^{-1} as gypsum and 3 kg Zn ha^{-1} as zinc oxide only at the BAU location. At each site and season, rice seedlings (2–3 per hill) were transplanted at the spacing of 20 cm × 20 cm. The rice cultivar grown were BRRI dhan 48, BR 22 and BRRI dhan 28 for the *Aus*, *Aman*, and *Boro* seasons, respectively, at BAU. Similarly, rice varieties grown at the BRRI site were BRRI dhan 43, BRRI dhan 46 and BRRI dhan 28 during the *Aus*, *Aman*, and *Boro* seasons, respectively.

All plots remained continuously flooded until two weeks before harvesting (Fig. S1). All other agronomic practices were followed as recommended.

2.4. Automated continuous measurement of N_2O and NO emissions

The automated closed chamber technique was used to collect air samples to measure N_2O and NO fluxes. The automated gas sampling and analysis system included four components: (i) automated gas chambers with compressed air-driven lids, (ii) the automated gas sampling unit, (iii) online gas analysis system and (iv) a control and data-logging unit (Fig. S2).

Twelve automated plexiglass chambers designed by the International Fertilizer Development Center (Gaihre et al., 2014) were installed in each location. Out of 12 chambers, nine chambers were installed under the continuous standing water (CSW) plots (three treatments, three replications) and three chambers were installed under AWD non-replicated plots (not reported in this paper). Chambers were placed between two rows of rice, leaving four border rows. Each chamber included an aluminum base (inner dimension, 118.8 cm × 12.5 cm) and aboveground plexiglass (5 mm thick) chamber (120.5 cm × 15.4 cm × 31.2 cm length, breadth and height, respectively). The aboveground chamber included a chamber lid, rubber gasket for proper sealing of the chamber during gas sampling time, two compressed air driven cylinders for opening and closing of the lid, DC fan for uniform mixing of air inside chamber and outer protective net. When the chamber was installed over the aluminum base, it covered a surface area of 0.148 m^2 and headspace volume of 0.0578 m^3 (57.8 L).

Sealing of the closed lid was achieved by a rubber gasket, while chamber base and chamber were sealed with water and a rubber gasket. Within a three-hour cycle, a chamber was closed for 40 min only during gas sampling. The lids of the chambers were opened and closed automatically with compressed air driven pneumatic pistons. The compressed air was supplied to the cylinders equipped with pneumatic pistons via nylon tubes (1/4″ outer diameter). Each nylon tube from a chamber was connected to a 12-port compressed air manifold. The manifold had 12 Clippard valves. Opening/closing of the valves and the chamber lids was controlled by the datalogger via relay controller (Fig. S2). Each chamber was fitted with two temperature sensors to monitor soil (5 cm depth) and air temperature and with one sensor for soil water potential (5–7 cm depth).

Six air samples were taken at eight-minute intervals (0, 8, 16, 24, 32, and 40 min) for 40 min at every three-hour sampling sequence. The zero-minute sample taken with open chamber represented the ambient air. The automated gas sampling system was running 24 h a day, seven days a week throughout the crop growing and fallow seasons repeating a three-hour sequence. There were three sets of four chambers, one set (one replication), i.e., four chambers were sampled in a one-hour cycle. The three-hour sequence consisted of three one-hour sequences that were identical, except the first hour was applied to chambers 1–4 (Rep 1), the second hour was applied to chambers 5–8 (Rep 2) and the third hour was applied to chambers 9–12 (Rep 3). Chambers 4, 8, and 12 represent the non-replicated treatments of the AWD experiment. Measurements done from AWD plots are not reported herein.

Gas samples collected from the chambers were passed to the analyzers using a Teflon tube with 1/4″ diameter. Each chamber was connected to a 13-port sample manifold (equipped with solenoid valve, i.e., KIP valves) with 1/4″ diameter Teflon tube (for gas sampling). Out of 13 valves in a sample manifold, 12 were used for air samples that came from the respective 12 chambers, while the last one was used for the calibration gases (i.e., connected to calibrator). Each sample valve in the manifold was controlled by the datalogger (Campbell Scientific, CR3000) via 16 port (channel) relay controller (SDMCD16 AC/DC Relay Controller). A total of 16 chambers could be connected to the relay; however, in this study, only 12 chambers were used. A vacuum pump either internal (fitted inside the gas analyzer, N_2O analyzer) or

Table 1
Physicochemical properties of soil used in the experiments.

Soil property	BAU	BRRI
pH–H_2O	5.5	6.2
Organic carbon (%)	1.76	1.18
Total N (%)	0.16	0.17
Available P (mg/kg)	2.97	9.84
Available K ($cmol_c$ kg^{-1})	0.09	0.10
Available S (mg/kg)	12	–
Particle size (%)		
Sand	11.44	29.96
Silt	72.4	40.10
Clay	16.16	29.94

external (fitted in the exhaust of gas analyzer, NO analyzer) circulates air continuously from the chamber headspace into the analyzers via sample manifold. The sample flow rate is determined by the vacuum pump and flow controller of the respective gas analyzers. The sample flow rate for NO and N_2O measurements were ca. 450 and ca. 750 $cm^3\ min^{-1}$, respectively. Air samples were extracted continuously from corresponding chambers for 2 min. After 2 min, the particular valve in the sample manifold closes and opens another one and continues accordingly following the sampling scheme. Air samples were filtered and dried before passing to the respective analyzers.

N_2O concentration of the air sample was measured by a Teledyne API (Advanced Pollution Instrumentation, USA) T320U Gas Filter Correlation analyzer. The T320U uses an infrared (IR) absorption principle. NO concentration of the air sample was measured by a Teledyne API (Advanced Pollution Instrumentation, USA) T200 Nitrogen Oxide analyzer. The T200 uses a chemiluminescent detection principle. It calculates the amount of NO present in an air sample by measuring the amount of chemiluminescence given off when a sample gas is exposed to ozone (O_3). Each air sample was analyzed continuously for 2 min, and the average of the last three measurements done in 15 s (one measurement every 5 s) was recorded in the datalogger. 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 concentrations, the high range values were used when concentrations of air sample exceeded the low range. The minimum detection limits of the flux measured based on the chambers used (57.8 L volume, 0.148 m^2 area, and 40-minute chamber closure time) were 17 and 4 $\mu g\ m^{-2}\ h^{-1}$ for N_2O and NO, respectively.

The N_2O and NO fluxes were calculated from the slope (positive or negative) of the linear model fitted to the concentration of either gas against the chamber closure time. An emission event was considered significant when the slope was significant at $P < 0.05$ and the R^2 value was 0.70 or higher. Therefore, flux rates when $R^2 < 0.7$ were not considered emission events and were discarded. The slope ($ppb\ min^{-1}$) from the significant emission events was corrected for air temperature, atmospheric pressure and the ratio of chamber volume to surface area using the following formula to determine the emission rate.

$$\text{Emission rate}\left(\mu g\ \text{NO–N or } N_2O\text{–N}\ m^{-2}\ h^{-1}\right) = \frac{ppb\ min^{-1} \times V \times MW \times 60}{\left[0.08206 \times (273 + T)^0K\right] \times A \times 1000}$$

where,

V is the volume of the gas chamber in L (57.8)
MW is the molecular weight of the respective gas in ng $nmol^{-1}$ (NO–N: 14, N_2O–N: 28)
60 is conversion factor for time ($min\ h^{-1}$)
0.08206 is the gas law constant ($L\ atm\ mol^{-1}\ {}^0K^{-1}$)
T is the temperature inside the chamber (°C)
A is the area covered by chamber (m^2, 0.148) and
1000 is conversion factor for mass ($ng\ \mu g^{-1}$).

Total cumulative seasonal emissions (g N_2O–N or NO–N) were calculated by summing the hourly emission rates. The gas sampling, analysis and data recording were controlled by a datalogger and the Loggernet software. We refer to Gaihre et al. (2014) for details of the automated gas sampling and analysis.

2.5. Adjustment of N_2O and NO fluxes by rice yield and nitrogen rate

The recommended N rate for UDP in Bangladesh is 25–35% lower than the recommended rate for broadcast PU application. Likewise, the recommended N application rate during the *Boro* season is higher because of the higher rice yield potential. To overcome the confounding effects that may occur between urea application method and N rate and, similarly, the confounding effects of season and N rate, adjustments to the cumulative N_2O and NO emissions were applied. Yield-scaled N_2O and NO emissions (g N t^{-1} grain) were determined using the following equation (illustrated with the N_2O case):

$$\text{Yield-scaled } N_2O_{(tr)}\left(g\ N_2O\text{–N}\ t^{-1}\text{grain}\right) = N_2O_{(tr)}/\text{Yield}_{(tr)}$$

where $N_2O_{(tr)}$ is the cumulative area-scaled N_2O emission (g N_2O–N ha^{-1}) from treatment t and replication r, and $\text{Yield}_{(tr)}$ is the yield ($t\ ha^{-1}$) from treatment t and replication r.

The N_2O emission factor (EF) was calculated and expressed as a percentage of applied N using the following equation:

$$N_2O\ EF(\%) = \left[N_2O_{(tr)} – N_2O_{(cr)}\right] * 100/\text{N Rate}_{(tr)}$$

where $N_2O_{(cr)}$ is the cumulative area-scaled N_2O emission (g N ha^{-1}) from control and replication r and $\text{N Rate}_{(tr)}$ is the N applied (g N ha^{-1}) to treatment t and replication r.

Urea briquettes were deep-placed in alternate rice rows, while the chamber was placed on the fertilized row. Deep placed N in a reduced zone becomes stable and tends to accumulate at the placement site as ammonium–N due to very slow spatial (horizontal) movement (Savant and de Datta, 1980; Savant and Stangel, 1990). Emissions from fertilized row may not be representative for unfertilized row. Therefore, emissions from unfertilized row were assumed to be similar with the emissions from control treatment. Then, adjusted emission factor for urea briquette was calculated from the average emissions of fertilized and unfertilized (control) rows using above equation.

2.6. Data analysis

The analysis of variance (ANOVA) was performed with a Generalized Linear Mixed Model (Gbur et al., 2012) following a split–split plot structure where the main plot was represented by the site, the subplot by the season and the sub-subplot by the treatment. Based on normality and variance homogeneity tests for the errors, the normal distribution was used as the probability distribution. A non-homogenous variance–covariance matrix was created for the estimation of parameters in the generalized linear mixed model. Standard errors were also calculated from the variance–covariance matrix to be used in mean comparisons. Site, season, treatment and the interactions from these three factors were handled as fixed effects. The interaction effects Treatment * Season, Treatment * Site and Treatment * Site * Season were tested with the error terms Replication * Treatment * Season, Replication * Treatment * Site and Replication * Treatment * Season * Site, respectively. These error terms were considered as random effect.

Degrees of freedom (DF) for the denominator (Den DF) in the calculation of the F statistic for all the fixed effects in the ANOVA were obtained using Kenward–Roger method (Gbur et al., 2012; Kenward and Roger, 1997). This methodology is applied to estimate appropriate DF for F calculation from the experiments that otherwise would not yield enough DF for the error terms.

All pairwise comparisons of treatment means from either significant interactions or simple effects were performed with the Tukey–Kramer test.

The ANOVA was applied to the seasonal cumulative N_2O and NO emissions, yield-scaled N_2O and NO emissions, the N_2O and NO emission factors and the N_2O and NO adjusted emission factors.

3. Results

3.1. Seasonal dynamics of N_2O fluxes: Aus–Aman 2013

Dynamics of N_2O fluxes measured hourly from all the treatments over *Aus* and *Aman* rice-growing seasons at BAU and BRRI are displayed in Fig. 2. N_2O fluxes were sporadic and event specific, which were likely reactions to the fertilizer and water management components of treatments. Peaks in fluxes were observed after topdressing of PU, during drying period and after reflooding of the dry soil for land preparation to next crop. The rest of the time during rice-growing season, N_2O fluxes showed similar patterns within a range of ± 20 μg N m^{-2} h^{-1} irrespective of treatments, seasons and location. Overall, there were high temporal and spatial variations in N_2O fluxes, particularly after topdressing of PU and during dry fallow.

At the BAU site, peaks in N_2O fluxes were observed in broadcast PU treatments after 2–7 days of topdressing, which were more prominent during *Aman* season than during *Aus* season (Fig. 2a). On the other hand, emission peaks were not observed in UDP treatments, resulting in similar emission patterns between control and UDP treatments. Occasional negative fluxes were observed in all treatments; however, they did not follow any consistent pattern. Over the rice-growing season, N_2O fluxes ranged from -13 to 23 μg N m^{-2} h^{-1} during *Aus* season, while they ranged from -18 to 90 μg N m^{-2} h^{-1} during *Aman* season.

At the BRRI site, N_2O emission peaks were observed after final drainage for harvest. As in BAU, fertilizer induced emission peaks in broadcast PU treatments were prominent during *Aman* season compared with *Aus* season. N_2O emission peaks observed during dry period (including fallow) were higher than fertilizer-induced peaks (Fig. 2b). Overall, N_2O fluxes ranged from -16 to 120 μg N m^{-2} h^{-1} during *Aus* season, while they ranged from -20 to 122 μg N m^{-2} h^{-1} during *Aman* season.

3.2. Seasonal dynamics of N_2O fluxes: Boro 2014

Dynamics of N_2O fluxes during *Boro* season for BAU and BRRI sites are shown in Fig. 3. As with *Aus–Aman* seasons, N_2O fluxes were affected by fertilizer treatments. Peak N_2O fluxes were observed after topdressing of PU. Magnitudes of fluxes were declining (BAU) or negligible (BRRI) with subsequent topdressing. Except during peak emission events, N_2O fluxes were within range of ± 20 μg N m^{-2} h^{-1}. Generally, magnitudes of N_2O fluxes in broadcast PU were higher in *Boro* (BAU) (Fig. 3a) compared with *Aus* and *Aman* seasons (Fig. 2a and b) while they were similar at BRRI. Nevertheless, fluxes from UDP treatments were similar to the control treatments during the *Boro* season as they were in the *Aus* and *Aman* seasons.

At the BAU site, fertilizer-induced emission peaks appeared 5–7 days after application (Fig. 3a), unlike 2–7 days after application in the *Aus–Aman* seasons (Fig. 2a). Moreover, emissions continued for a longer time (one week) compared with the *Aus–Aman* seasons (2–3 days). Emission peaks in the UDP treatment were observed only once after 5–7 days of deep placement. Nevertheless, the emission peak was small compared with broadcast PU. For the rest of the period during the rice-growing season, the emission rates between control and UDP treatments were similar. Some emission peaks occurred after final drainage for harvest. N_2O fluxes varied from -13 to 269 μg N m^{-2} h^{-1}.

At the BRRI site, emission rates were relatively lower compared with the BAU site. Peaks in N_2O fluxes were prominent only after the first topdressing of PU, while they were very small to negligible after the second and third topdressing. Nevertheless, emission peaks were observed from both UDP and broadcast PU treatments after final drainage for harvest. Overall, N_2O fluxes varied from -16 to 106 μg N m^{-2} h^{-1}.

3.3. Seasonal dynamics of NO fluxes

Dynamics of NO fluxes measured hourly from all the treatments during three cropping seasons (*Aus* 2013, *Aman* 2013 and *Boro* 2014) at BAU and BRRI are displayed in Figs. S3 and S4. Occasional emission peaks appeared after broadcast PU or during the dry period. However, the effect of the N fertilizer treatments was not consistent on NO fluxes. Overall, NO fluxes were very low. Most of the times during rice-growing period, fluxes were below 5 μg N m^{-2} h^{-1} at BAU. Fluxes were relatively higher at the BRRI site in all cropping seasons compared with BAU. Moreover, the temporal variation in fluxes was also higher at BRRI. NO fluxes varied from 0 to 5 μg N m^{-2} h^{-1} at BAU, while it was from 0 to 25 μg N m^{-2} h^{-1} at BRRI. Emission patterns showed seasonal differences; they were relatively higher during the *Aman* and *Boro* seasons than that of the *Aus* season.

3.4. Cumulative N_2O emissions, yield-scaled N_2O emissions, and N_2O emission factor

The significant interactions of Treatment * Season and Treatment * Site from the ANOVA for the N_2O emissions (area-scaled in g N ha^{-1} $season^{-1}$), the yield-scaled N_2O emissions (g N t^{-1} grain)

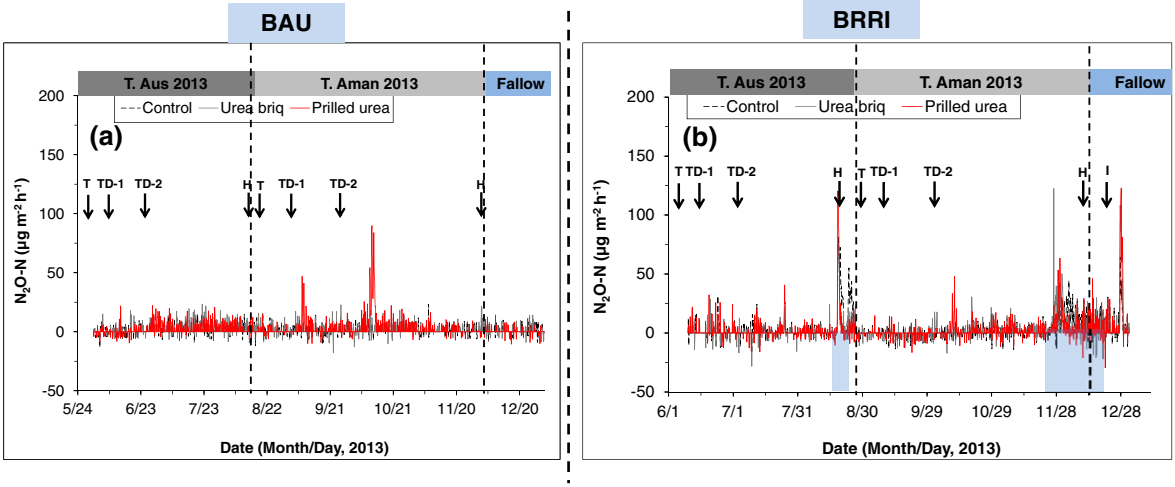

Fig. 2. Seasonal variations in N_2O emission rates under different fertilizer treatments during the *Aus*, *Aman* and Fallow seasons, 2013 at BAU and BRRI. T, TD-1, TD-2, H are transplanting, first and second topdressing and harvesting, respectively. Shaded area indicates the drying period.

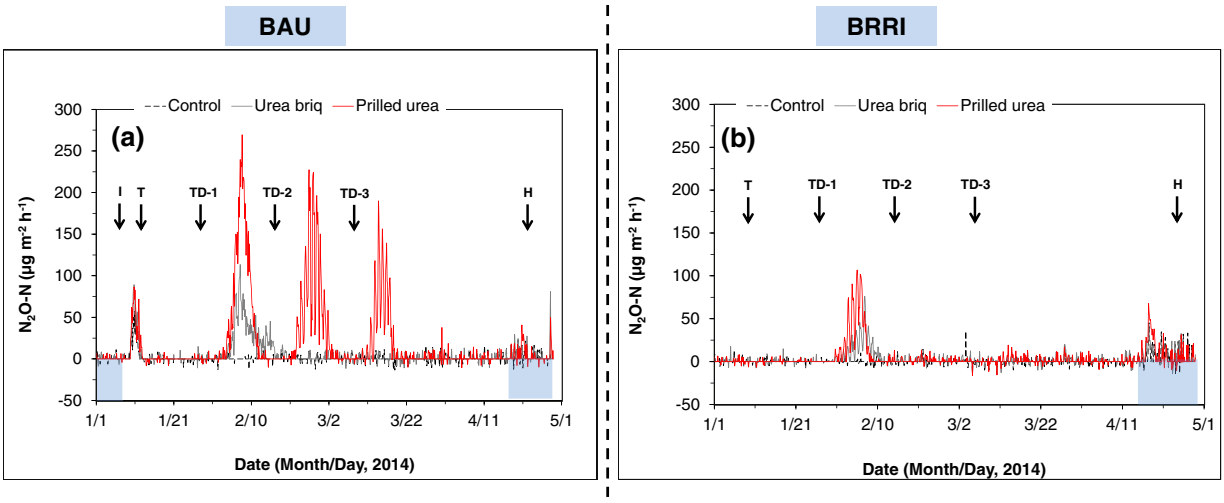

Fig. 3. Seasonal variations in N_2O emission rates under different fertilizer treatments during the *Boro* season, 2014 at BAU and BRRI. I, T, TD-1, TD-2, TD-3, H represent initial irrigation, transplanting, first, second and third topdressing and harvesting, respectively. Shaded area indicates the drying period.

and adjusted N_2O emission factor (%, w/w) (Table 2) point out separate significant effects of season and site over treatment. The N_2O emission factor, unadjusted for N rate inside the chambers, was significant only for the treatment effect in the ANOVA.

The mean comparisons in Table 3 indicate that, independent of the different methods of calculation, a significant reduction in N_2O emission occurred with the UDP. During the *Boro* season, UDP across both sites resulted in a significant cumulative N_2O emission reduction by about 61%, the yield-scaled N_2O emission reduction by about 67%, and the adjusted N_2O emission factor reduction by about 84% compared with broadcast PU application. Similarly, the UDP emitted significantly lower N_2O at the BAU site. The N_2O emission reductions due to UDP were 66%, 70%, and 82% for cumulative N_2O emissions, yield-scaled N_2O emissions, and adjusted N_2O emission factor, respectively.

The consistency exhibited by the three ways of estimating the N_2O emissions in both interactions, treatment by season and treatment by site, can be interpreted as evidence of unbiased estimations made by the cumulative (unadjusted) N_2O emission measurements in spite of apparent confounding effects of N rate and urea application method, or confounding effects of N rate and season.

3.5. Cumulative NO emissions, yield-scaled NO emissions, and NO emission factor

Of the four forms of presenting NO emissions–cumulative NO emission (area-scaled in g N ha^{-1} $season^{-1}$), yield-scaled NO emission (g N t^{-1} grain), NO emission factor (%, w/w), and adjusted NO emission factor (%, w/w) — only the yield-scaled NO emissions presented

Table 2
Analysis of variance for the different estimation forms of N_2O and NO emissions.

		N_2O–N emission (g ha^{-1})		Yield-scaled N_2O–N emission (g t^{-1} grain)		NO–N emission (g ha^{-1})		Yield-scaled NO–N emission (g t^{-1} grain)	
Effect	Num DF	Den DF	Pr > F	Den DF	Pr > F	Den DF	Pr > F	Den DF	Pr > F
Rep	2	15.75	0.137	10.23	0.191	11.33	0.058	8.39	0.046
Treatment (T)	2	15.05	<.001	19.63	0.001	14.92	0.694	16.75	<.001
Site	1	15.05	0.557	19.63	0.845	14.92	<.001	16.75	<.001
Season	2	18.93	<.001	17.78	<.001	10.11	<.001	10.52	<.001
Season * Site	2	18.93	0.365	17.78	0.458	10.11	<.001	10.52	<.001
T * Season[1]	4	20.6	0.001	19.28	0.009	10.76	0.334	11.21	0.003
T * Site[2]	2	15.05	0.017	19.63	0.001	14.92	0.877	16.75	0.105
T * Season * Site[3]	4	20.6	0.114	19.28	0.061	10.76	0.618	11.21	0.350

1: Tested with error term Rep * Trt * Season, 2: tested with error term Rep * Trt * Site
3: Tested with error term Rep * Trt * Season * Site

		N_2O–N emission factor (%, w/w)[4]		Adjusted N_2O–N emission factor (%, w/w)[5]		NO–N emission factor (%, w/w)[4]		Adjusted NO–N emission factor (%, w/w)[5]	
Effect	Num DF	Den DF	Pr > F	Den DF	Pr > F	Den DF	Pr > F	Den DF	Pr > F
Rep	2	3.1	0.361	5.0	0.42	9.3	0.222	6.39	0.053
Treatment (T)	1	15.3	0.040	12.9	0.00	12.4	0.650	22.00	0.439
Site	1	15.3	0.010	12.9	0.01	12.4	0.577	22.00	0.053
Season	2	9.7	0.003	9.6	<.01	11.5	0.003	11.95	0.049
Season * Site	2	9.7	0.164	9.6	0.09	11.5	0.257	11.95	0.042
T * Season[1]	2	9.7	0.125	9.6	0.01	11.5	0.765	11.95	0.147
T * Site[2]	1	15.3	0.217	12.9	0.03	12.4	0.867	22.00	0.430
T * Season * Site[3]	2	9.7	0.340	9.6	0.27	11.5	0.667	11.95	0.140

1: Tested with error term Rep * Trt * Season, 2: tested with error term Rep * Trt * Site
3: Tested with error term Rep * Trt * Season * Site, 4: calculated with formula: (emissions from fertilizer treatment-emissions from N0 treatment)/ N rate * 100
5: Calculated with adjusted average emissions (average emissions of fertilized row and unfertilized row) for urea briquette

Table 3
Pairwise comparison of means for emission forms that presented significance (P ≤ 0.05) for TRT or interactions involving TRT in the analysis of variance.

Season	Site	Trt	N_2O–N emission (g ha^{-1})	Yield-scaled N_2O–N emission (g t^{-1} grain)	N_2O–N emission factor (%, w/w)	Adjusted N_2O–N emission factor (%, w/w)	Yield-scaled NO–N emission (g t^{-1} grain)
Aus 2013	All	Control	44.13a	14.98a			0.779a
		Prilled urea	43.30a	10.80a	−0.001a	−0.001a	0.591a
		Urea briquette	37.97a	9.01a	−0.012a	−0.006a	0.567a
Aman 2013	All	Control	32.18a	11.70a			3.426a
		Prilled urea	66.82a	18.63a	0.044a	0.044a	2.697a
		Urea briquette	47.90a	12.05a	0.030a	0.015a	2.890a
Boro 2014	All	Control	66.91b	32.76b			5.118a
		Prilled urea	411.56a	81.92a	0.331a	0.331a	1.562b
		Urea briquette	158.96b	26.81b	0.118a	0.059b	0.968b
All	BAU	Control	22.77b	9.30b			
		Prilled urea	221.84a	48.39a	0.200a	0.200a	
		Urea briquette	75.63b	13.97b	0.074a	0.037b	
All	BRRI	Control	72.70a	30.33a			
		Prilled urea	125.96a	25.84a	0.050a	0.050a	
		Urea briquette	87.59a	17.95a	0.016a	0.008a	
All	All	Control					
		Prilled urea			0.120a		
		Urea briquette			0.045b		

Within a season or site, means followed by the same letters are not significantly different at P ≤ 0.05 (P values adjusted by the Tukey–Kramer method), urea briquette and prilled urea represent UDP and broadcast, respectively.

significance with respect to treatment, as the Treatment * Season interaction (Table 2). The means from the interaction Treatment * Season for yield-scaled NO emissions in Table 3 show overall higher emissions from control plots than urea plots. The control plots had significantly higher yield-scaled NO emissions during the *Boro* season across both sites.

Significant Season * Site interaction was observed with the cumulative NO emissions and the adjusted NO emission factor (Table 2). At the BAU site, during the *Boro* season NO emissions were significantly higher (>70% higher) than the other two seasons (Table 4). At the BRRI site, significant differences were observed among all seasons. During *Aman*, NO emissions were significantly higher at ~47% and ~84% compared with *Boro* and *Aus* seasons, respectively, while the emissions were significantly higher by 69% during *Boro* compared with the *Aus* season. Also, for the three seasons, the BRRI site consistently presented significantly higher NO emissions than the BAU site: ~56% higher in *Aus*, ~92% higher in *Aman*, and ~48% higher in *Boro*. Means from the adjusted NO emission factor presented few similarities with the NO emission pattern: the adjusted NO emission factor was significantly higher at BRRI than at BAU during the *Boro* season, and significantly higher during *Boro* than the *Aus* season at the BRRI site. In contrast, the unadjusted NO emission factor showed significantly higher emissions during the *Aman* season across sites. The negative mean values for N_2O and NO emission factors (Tables 3 and 4) were associated with higher emissions from the control plots than the urea treatments.

4. Discussion

4.1. Effects on N_2O emissions

This study reveals that UDP has a large effect on N_2O emissions. UDP at 7–10 cm depth reduced N_2O emissions ranging from 61% to 84% relative to broadcast PU depending on the type of N_2O emission assessment used (Table 3). It is evident that production of N_2O mainly occurs near the soil surface (Venterea and Stanenas, 2008; Yoh et al., 1997) due to higher microbial nitrification and denitrification compared with deeper soil layers. Previous studies have shown that the placement of urea briquettes at the reduced zone (7–10 cm depth) retains N in NH_4^+ form. The diffusion of NH_4^+–N from a reduced zone is very slow, and a negligible amount of NH_4^+–N comes to the surface of the soil or in floodwater (Chien et al., 2009; Kapoor et al., 2008; Rochette et al., 2013; Savant and Stangel, 1990). Thus, UDP simply decreases the supply of inorganic N substrates within the most biologically active zone where they can be converted to N_2O via nitrification and/or denitrification (van Kessel et al., 2013). On the other hand, the peaks in N_2O emissions after broadcast PU could be associated with the conversion of NH_4^+–N, that

Table 4
Pairwise comparison of means for significant season, site, or Season * Site effects from the ANOVA of different types of NO emissions.

Season * Site interactions				Season effect		
Season	Site	NO–N emission (g ha^{-1})	Adjusted NO–N emission factor (%, w/w)[a]	Season	Site	NO–N emission factor (%, w/w)
Aus 2013	BAU	1.4353b	−0.00035a	*Aus* 2013	All	−0.00017b
	BRRI	3.2509a	0.00008a			
Aman 2013	BAU	1.6017b	0.00058a	*Aman* 2013		0.00296a
	BRRI	19.7503a	0.00334a			
Boro 2014	BAU	5.4211b	−0.00334b	*Boro* 2014		−0.00403b
	BRRI	10.4948a	0.00360a			
Aus 2013	BAU	1.4353b	−0.00035a			
Aman 2013		1.6017b	0.00058a			
Boro 2014		5.4211a	−0.00145a			
Aus 2013	BRRI	3.2509c	0.00008a			
Aman 2013		19.7503a	0.00334a, b			
Boro 2014		10.4948b	−0.00361b			

Within a season or site, means sharing with the same letter do not have significant differences between them.
[a] Calculated with adjusted average emissions (average of fertilized row and unfertilized row) for urea briquette.

arise from urea hydrolysis, to NO_3^-, i.e., nitrification and subsequent loss of NO_3^- by denitrification. The peaks in N_2O flux observed after broadcast PU application are consistent with previous studies (Bronson et al., 1997a; Chen et al., 1997; Sander et al., 2014). The peaks in N_2O flux were rarely observed in UDP plots at both locations. The small emission peaks which occurred in *Boro* season could also be minimized with proper placement of urea briquettes in reduced zone and closing with soil. Therefore, this study supports the hypothesis that UDP (7–10 cm depth) can be an effective strategy for mitigating N_2O emissions in continuously flooded conditions. Based on a review study, van Kessel et al. (2013) made similar conclusions, particularly under no tillage or reduced tillage condition.

High temporal (within and between growing seasons) and spatial variations (within treatments and between sites) of N_2O fluxes observed in this study (Tables 2 and 3, Figs. 2 and 3) are consistent with previous studies (Akiyama et al., 2005; Liang et al., 2013; Scheer et al., 2012). The emission peaks after topdressing of PU were not consistent. Though some studies (Sander et al., 2014) reported a decreasing tendency of N_2O flux during succeeding split applications of N because of increasing competition of rice plants with nitrifying organisms for inorganic N formed from hydrolysis of the applied urea, this study did not show a clear trend in that respect. Nevertheless, relatively higher N_2O emission rates were observed after the first topdressing compared with the second and third topdressing, particularly during the *Boro* season, which yielded significantly higher emissions from broadcast PU compared with UDP (Table 3). Negligible emission of N_2O after the second and third N applications (Fig. 3) could be due to rapid uptake of inorganic N by rice plants before it nitrified or denitrified (Sander et al., 2014). UDP application during the *Boro* season and at the BAU site resulted in significant N_2O emission reductions with respect to PU. The *Boro* season is characterized by higher N application rates, lower volatilization losses due to lower temperatures and controlled irrigation, and the latter also resulting in reduced N runoff losses. All of these result in potentially higher soil N for nitrification and denitrification from PU application. Soil and environmental conditions that reduce N losses due to volatilization and runoff from PU application at the BAU site may explain the significantly higher N_2O emissions compared with UDP.

The emission of N_2O is not solely driven by the source, amount and methods of N-fertilizer applied (IPCC, Tier 1), but it is also dependent on the overall performance of the cropping system; i.e., N uptake and crop yield related to N fertilizer applied (van Kessel et al., 2013), water management, agro-climate, yield potential and soil properties. It is also reported that the N losses, including N_2O emissions, increases when N is applied in excess, i.e., beyond plant uptake (Kim et al., 2013b; Shcherbak et al., 2014). However, in this study, N input for both fertilizer treatments were optimum. Therefore, higher emissions from PU (*Boro* season, BAU site) compared with UDP might be due to the effects of application method rather than the differences in N rate between the two treatments. This is also evident from emission factor and yield-scaled emissions (Tables 2 and 3). The N rate for UDP was 25% (*Aus-Aman*) to 33% (*Boro*) lower than in PU when we consider N supply to entire experimental plots, but N rate in UDP inside the gas chamber was two times higher than PU. This is because chambers in the UDP plots were placed only in rows, where urea briquettes were deep placed. The adjusted emission factor accounts for the higher N rate. More measurements are needed using a chamber that covers two rows of rice or by placing two chambers in a plot, one in the fertilized and another in the unfertilized row.

Occasional negative fluxes observed in this study are in line with several previous studies. Meta-analyses (Chapuis-Lardy et al., 2007; Kim et al., 2013a) showed that agricultural soil could act as an N_2O sink. Therefore, negative N_2O fluxes measured in this study were not ignored. However, all the non-significant fluxes ($R^2 < 0.7$, $p > 0.05$) were ignored. Several other field measurement studies in flooded rice soils have also recorded negative fluxes, which could be associated with the reduction of N_2O to N_2 (Firestone and Davidson, 1989). Negative fluxes due to N_2O consumption or uptake tend to occur under conditions of low soil inorganic N and high soil water contents (Chapuis-Lardy et al., 2007).

As observed in this study, N_2O emissions are generally low in continuously flooded rice systems (Bronson et al., 1997a; Chen et al., 1997; Ma et al., 2009). Therefore, negligible emissions occur throughout the rice-cropping season except some emission peaks after N fertilizer application (broadcast PU). However, occasional emission peaks appear when the rice field is drained and during dry fallow due to increased nitrification during the drying period and denitrification after the reflooding of the dry soil (Bronson et al., 1997b). Nevertheless, the high emission peaks are generally limited to a few hours to days of each event such as flooding, drying or N fertilizer application; thereafter, emissions are negligible. Our results confirm that the N_2O emissions from flooded rice fields are generally sporadic and event-specific; i.e., emissions occur after broadcast PU, during wetting and drying.

Management of N and irrigation water can influence production of both N_2O and NO by affecting microbial nitrification and denitrification. These results imply a potential of UDP technology for mitigating N_2O in lowland rice cultivation. Since UDP also significantly reduces N losses, particularly NH_3 volatilization (Rochette et al., 2013) while saving fertilizer N (urea), reducing weed populations and increasing crop productivity (Mohanty et al., 1999), it could be an effective climate smart technology.

4.2. NO emissions

NO is produced during both nitrification and denitrification. However, pronounced emissions are observed in upland crops (Akiyama and Tsuruta, 2002; Hou et al., 2010; Zhou et al., 2010) compared with lowland rice fields (Zhou et al., 2010). Zhou et al. (2010) and Bible et al. (in preparation) 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 that accounted for 85% of the total NO emissions. Significant NO fluxes from upland crop and during the dry period in rice fields suggest that nitrification contributes more to NO emissions compared with denitrification. Likewise, N_2O is a more dominant substrate for denitrification than NO. However, during denitrification, NO could further be reduced to N_2 as discussed earlier. Hou et al. (2010) also observed NO production only in those sites where urea was placed, and most of the produced NO was consumed instead of diffusing away from the production site in soils. In this study, experimental plots were flooded most of the time. Therefore, cumulative seasonal total N loss as NO emissions was negligible.

It is widely reported that N fertilizer has a direct influence on NO fluxes. Distinct emission peaks could be observed after N fertilizer application in both upland (Akiyama and Tsuruta, 2002; Zhao et al., 2015) and lowland crop fields (Zhou et al., 2010). In this study, however, the emission peaks observed after fertilizer application were very small and not consistent. The fertilizer induced EF in this study is extremely low compared with 0.7% of applied N (global mean) reported by Bouwman et al. (2002) and 0.04% of rice fields in China (Huang and Li, 2014). Nevertheless, experiments conducted by Hou et al. (2010) in upland crop (Chinese cabbage) suggest that UDP is highly effective in reducing NO emissions. In this study, only the yield-scaled NO emissions had significant fertilizer treatment effects with respect to Treatment * Season interaction, with significantly higher NO emissions from control plots during the *Boro* season across both sites.

However, there was significant interaction of season and site, with NO emissions significantly higher during the *Boro* season at BAU, and there were also significant differences among *Aus* > *Boro* > *Aman* at BRRI (Table 4). During all three seasons, NO emissions were significantly higher at BRRI than at BAU. The higher emissions at BRRI may be attributed to higher NO background (urban area) and higher soil

drainage. The seasonal differences may be attributed to longer growth duration during the *Boro* and *Aman* seasons with respect to the *Aus* season. Both broadcast PU and UDP had higher emissions than control during the *Aman* season. NO emissions could be controlled by soil type and other environmental factors, such as temperature, in addition to N fertilizer and water management (Pang et al., 2009).

5. Conclusions

Significant N_2O emission peaks were observed after each broadcast application of PU irrespective of the rice-growing seasons, while emission peaks were rarely observed after deep placement of urea briquettes. Cumulative seasonal total emissions under continuously flooded conditions were mostly contributed by fertilizer induced emission peaks. Regardless of the growing season, N_2O emissions from broadcast PU were almost two to three times higher compared with UDP. Moreover, the effect of broadcast PU was site- and season-specific; N_2O emissions were significantly higher at the BAU site compared with BRRI and higher during the *Boro* season compared with the *Aus* and *Aman* seasons, probably due to differences in soil and climatic factors. On the other hand, the effect of UDP was consistent irrespective of the growing seasons and sites. Emissions from UDP were not statistically different with the control treatment.

The NO emissions were very small compared with N_2O emissions during all rice-growing seasons. Emission peaks after broadcast PU and during the dry period were not consistent. Significantly higher yield-scaled NO emissions were observed from control plots during the *Boro* season across both sites. NO emissions showed significant spatial and seasonal variations with higher emissions at BRRI site compared with BAU during all three seasons, while the *Aus* season emissions were significantly lower at both the locations. UDP, in addition to saving N fertilizer and increasing crop productivity (Kapoor et al., 2008), drastically reduces N losses as ammonia volatilization (Rochette et al., 2013) and N_2O emissions. Therefore, UDP could be considered as a climate smart technology.

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