# Methane and Nitrous Oxide Emissions from Flooded Rice Systems following the End-of-Season Drain

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#### Abstract

Large CH, and N<sub>2</sub>O fluxes can occur from flooded rice (Oryza sativa L.) systems following end-of-season drainage, which contribute significantly to the total growing-season greenhouse gas (GHG) emissions. Field and laboratory studies were conducted to determine under what soil water conditions these emissions occur. In three field studies, GHG fluxes and dissolved CH, in the soil pore water were measured before and after drainage. Across all fields, approximately 10% of the total seasonal CH<sub>4</sub> emissions and 27% of the total seasonal N<sub>2</sub>O emissions occurred following the final drain, confirming the importance of quantifying postdrainage CH<sub>4</sub> and N<sub>2</sub>O emissions. Preplant fertilizer N had no effect on CH<sub>4</sub> emissions or dissolved CH<sub>4</sub>; however, increased postdrainage N<sub>2</sub>O fluxes were observed at higher N rates. To determine when postdrainage sampling needs to take place, our laboratory incubation study measured CH, and N<sub>2</sub>O fluxes from intact soil cores from these fields as the soil dried. Across fields, maximum CH<sub>4</sub> emissions occurred at approximately 88% waterfilled pore space (WFPS), but emissions were observed between 47 and 156% WFPS. In contrast, maximum N<sub>2</sub>O emissions occurred between 45 and 71% WFPS and were observed between 16 and 109% WFPS. For all fields, gas samplings between 76 and 100% WFPS for CH<sub>4</sub> emissions and between 43 and 78% WFPS for N<sub>2</sub>O emissions was necessary to capture 95% of these postdrainage emissions. We recommend that frequent gas sampling following drainage be included in the GHG protocol of total GHG emissions.

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J. Environ. Qual. 44:1071–1079 (2015) doi:10.2134/jeq2014.11.0497 Received 24 Nov. 2014. Accepted 12 May 2015. \*Corresponding author (aaadvientoborbe@ucdavis.edu). GRICULTURE is the major source of current global  $CH_4$  and  $N_2O$  emissions, contributing about 60 and 58% of total anthropogenic emissions of  $CH_4$  and  $N_2O$ , respectively (Ciais et al., 2013). With regard to  $CH_4$ , rice (*Oryza sativa* L.) production remains the largest emission source from a single sector and accounts for 18% of total agricultural  $CH_4$  emissions (Ciais et al., 2013). Although rice systems also emit  $N_2O$ ,  $CH_4$  emissions from rice make up approximately 90% of the global warming potential in flooded rice systems (Linquist et al., 2012b).

Most studies evaluating  $CH_4$  emissions in rice systems show that the majority of emissions occur during the growing season when the fields are flooded. However, significant  $CH_4$  and  $N_2O$ fluxes have also been observed following drainage at the end of the season (Wassmann et al., 2002; Zou et al., 2005). Therefore, to obtain accurate total seasonal greenhouse gas (GHG) emissions from rice fields, it is necessary to measure GHG emissions during the entire season including flooded and nonflooded periods.

In most rice systems, fields remain flooded until a few weeks before harvest. Drainage and the subsequent soil drying reduce  $CH_4$  production as the soil redox potential (Eh) increases (Sigren et al., 1997). However, entrapped  $CH_4$  can be released from the soil after drainage (Wassmann et al., 2002). Denier van der Gon et al. (1996) reported that between 6 and 14% of the total seasonal  $CH_4$  emissions were released following drainage. This release of entrapped  $CH_4$  was corroborated by other GHG studies in flooded rice systems (Yagi et al., 1996; Wassmann et al., 2000; Adviento-Borbe et al., 2013; Pittelkow et al., 2013; Liang et al., 2013).

Nitrous oxide emissions depend on the presence of water logging, soil Eh, and the amount of N inputs applied (Cai et al., 1997; Zou et al., 2005). Emissions of  $N_2O$  during field drainage may range from 10 to 75% of the total seasonal  $N_2O$  emissions (Zou et al., 2005; Zhao et al., 2011; Adviento-Borbe et al., 2013). In flooded rice systems,  $N_2O$  emissions are not normally observed because of low Eh, and most N gas is released as  $N_2$  (Mosier et al.,

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**Abbreviations**: Eh, redox potential; GC, gas chromatograph; GHG, greenhouse gas; MC, soil water content;  $MC_{v}$ , soil volumetric water content; PVC, polyvinyl chloride pipe; WFPS, water-filled pore space.

1990; Hou et al., 2000). Instead, the majority of  $N_2O$  emissions in rice systems occur during drainage periods when soils change from anaerobic to aerobic conditions (Towprayoon et al., 2005; Yao et al., 2012; Adviento-Borbe et al., 2013). Furthermore, long-term exposure to  $O_2$  during soil drainage increases  $N_2O$ emissions because soil Eh increases, promoting nitrification (Reddy and Patrick, 1975; Yu et al., 2004). The optimal soil water content for nitrification activity has been reported at 60% waterfilled pore space (WFPS) (Linn and Doran, 1984). Hou et al. (2012) reported a range of 78 to 85% WFPS for  $N_2O$  emissions from controlled irrigated rice paddies.

Importantly, some have not observed postdrainage  $CH_4$ and  $N_2O$  emissions from rice systems (Setyanto et al., 2000). This may be due to low  $CH_4$  concentrations in the reduced soil layer before the drainage event (Wassmann et al., 2002) or low amounts of N substrates (i.e.,  $NO_3^{-}$ ) remaining in the field (Yan et al., 2000). Furthermore, it should also be considered that often gas emissions following drainage are not measured (Yagi and Minami, 1990; Lindau, 1994; Xie et al., 2010) or gas samplings were set at weekly intervals resulting in these emissions not being measured (Wassmann et al., 1994; Cai et al., 1997; Liu et al., 2010).

It remains important to determine GHG emissions following drainage to fully quantify seasonal GHG emissions. Identifying the critical range in soil water content when these postdrainage  $CH_4$  and  $N_2O$  emissions occur is necessary to sample at the correct times. Therefore, the objective of this study was to quantify postdrainage emissions of  $CH_4$  and  $N_2O$  and to determine the soil water conditions resulting in these postdrainage emissions.

# **Materials and Methods**

## **Field Experiments**

Three field experiments at separate locations in California were conducted in 2011 to quantify  $CH_4$  and  $N_2O$  emissions in rice systems. All three field trials were arranged in randomized complete block design with three blocks. Results and details from these experiments have been reported elsewhere (Field A,

Table 1. Location and properties of the three field soils used in this study.

Simmonds et al., 2015; Field B, Adviento-Borbe et al., 2013; Field C, Pittelkow et al., 2013). For Field A, which examined varietal effects on GHG emissions, only the variety M-206 at N rate of 130 kg N ha<sup>-1</sup> was used for this study. In Fields B and C, the experiments evaluated the effects of N rate on seasonal CH<sub>4</sub> and N<sub>2</sub>O emissions and N rates of 0, 100, and 140 kg N ha<sup>-1</sup> from Field B and 0, 200, and 260 kg N ha<sup>-1</sup> from Field C were used here (Table 1). Soil samples from each experimental plot were taken as described below for a laboratory incubation study.

#### Field Measurements of Methane and Nitrous Oxide Emissions

In all fields, growing-season CH4 and N2O emissions were measured daily during N fertilization, flooding, and drainage events and weekly during the season in each treatment plot using a static vented-chamber technique (Hutchinson and Livingston, 1993). Briefly, vented flux chambers consisted of a chamber base (29.5 cm in diameter and 22.9 cm in height), a chamber lid (7.6cm tall), and chamber extension (15.3–80.6 cm to accommodate growing rice plant inside the chamber), all made of polyvinyl chloride pipe (PVC). The chamber base was placed 15 cm into the soil to provide a solid foundation during gas sampling and left in place during the entire cropping. To prevent water and root restriction, two holes (2.86-cm diameter) were drilled on upper sides of the base and four 11-cm diameter holes in the bottom of the base. A 12-V fan installed inside the chamber lid (Allied Electronics) was used to mix the headspace gas for 1 min before sampling. Chamber air temperature was measured using a thermocouple wire. Gas sampling occurred between 0900 and 1200 h, and the sequence of gas measurements in the treatment plots was randomized to avoid bias caused by changing air temperature. Twenty-five-milliliter gas samples were taken from the chamber at four equal time intervals (21 min) within an hour of chamber closure and stored in a pre-evacuated 12-mL glass vials (Labco Ltd.) with rubber septa double sealed with 100% silicon before gas analysis. Soil trace gases were analyzed on a GC 2014 gas chromatograph (GC) (Shimadzu Scientific) with a <sup>63</sup>Ni electron capture detector set at 325°C for N<sub>2</sub>O concentrations and flame ionization detector set at 250°C for

Properties†	Field A	Field B	Field C
Location	Rice Experiment Station, Biggs, CA	Commercial rice field, Robbins, CA	Commercial rice field, Arbuckle, CA
Latitude	39.46°	39.01°	39°
Longitude	121.73°	121.70°	121.91°
Elevation, m	30	11	8
Soil classification	Fine, smectitic, thermic Xeric Epiaquerts and Duraquerts	Fine, smectitic, thermic, Typic Argixerolls	Fine, smectitic, thermic Xeric Endoaquerts
Soil	Esquon–Neerdobe clay	Marcum clay loam	Clear Lake clay
Soil fraction			
Sand, %	23	30	9
Silt, %	30	42	35
Clay, %	47	28	56
Chemical properties			
рН	4.80	5.46	6.43
Electrical conductivity, dS m <sup>-1</sup>	0.19	0.17	0.38
Cation exchange capacity, $cmol_c kg^{-1}$	33.8	24.7	48.8
Total organic C, g kg <sup>-1</sup>	12.6	13.7	18.2
Total N, g kg⁻¹	0.77	1.1	1.5

† Soil properties represent 0- to 0.15-m soil depth.

 $CH_4$  concentrations. Methane and  $N_2O$  were separated by a stainless steel column packed with Hayesep D, 80/100 mesh at 75°C. A 1 mL headspace gas was injected into the GC inlet port using an autosampler (Bandolero, XYZTEK).

Fluxes of N<sub>2</sub>O and CH<sub>4</sub> were estimated from the linear increase of gas concentration over time based on  $r^2 \ge 0.90$  (Liu et al., 2010; Shang et al., 2011) while providing the maximum available flux data in the analysis of gas emissions. Gas concentrations were converted to mass per unit volume (g N<sub>2</sub>O or CH<sub>4</sub> L<sup>-1</sup>) using the Ideal Gas Law at chamber air temperature measured during each sampling event and 0.101 MPa. Fluxes of N<sub>2</sub>O and CH<sub>4</sub> were computed as follows:

$$F = \frac{\Delta C}{\Delta t} \times \frac{V}{A} \times \infty$$
[1]

where *F* is gas flux rate for N<sub>2</sub>O/CH<sub>4</sub> (g N<sub>2</sub>O-N/CH<sub>4</sub>-C ha<sup>-1</sup> d<sup>-1</sup>),  $\Delta C/\Delta t$  denotes the increase or decrease of gas concentration in the chamber (g L<sup>-1</sup> d<sup>-1</sup>), *V* is the chamber volume (L), *A* is the enclosed surface area (ha), and  $\propto$  is a conversion coefficient for elemental N and C (28/44 for N<sub>2</sub>O; 12/16 for CH<sub>4</sub>). A significance test was made to determine if the gas flux was close or equal to zero at *P* value < 0.05. Gas fluxes that failed the linearity test were not included in the data analysis and accounted for <3% of the total data set, while gas fluxes that failed significance and detection tests were set to zero flux. A complete discussion of chamber flux method is described in Adviento-Borbe et al. (2013).

#### **Dissolved Methane Gas Concentrations**

Concentrations of dissolved CH4 in soil solution were measured in the latter part of the season in each field on an approximately weekly basis and more frequently following the drainage. In Field A, dissolved CH<sub>4</sub> in soil solution was collected using a soil pore water sampler made from PVC (1.5 cm i.d.) and silicon tubing. The pore water sampler consisted of a perforated plastic irrigation coupler (0.38-cm diameter) with 0.2-cm diameter holes and a sealed end (i.e., perforated plastic pipette) inserted to a 10-cm depth. MasterFlex silicon tubing (0.08-cm diameter) connected the plastic coupler to a three-way stopcock attached to a 5-µm nylon filter from which the pore water samples were collected (McKee et al., 1988; Rejmankova and Post, 1996). The sampler could hold 2.5 mL of pore water. Since this pore water sampler did not provide a stable and air tight fit in the reduced soil layer because of the relatively large tube diameter, a porous ceramic tubing with 2.5 mm i.d. and 10-cm length (Rhizon soil moisture sampler; Rhizosphere Research Products) was used in fields B and C. For these fields, soil solutions were sampled from the same N-rate treatments where soil cores were obtained for the laboratory incubation using a porous ceramic tube. In each treatment, a Rhizon soil sampler was installed diagonally across 0- to 10-cm depth where it remained in place throughout the growing season. For both Rhizon and PVC pore water samplers, a pre-evacuated glass vial was connected to the sampler through a needle and collected 5 mL of soil solution. The soil solution was shaken for 60 sec and 2 mL of headspace gas was transferred to pre-evacuated glass vial (Alberto et al., 2000). The headspace gas was immediately diluted with 15-mL He gas and analyzed using a GC-2014, as described

above. The concentration of dissolved  $\rm CH_4$  was calculated as described by Alberto et al. (2000).

#### Measurements of Other Ancillary Data

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At 1 to 3 wk intervals, soil cores of 4.6-cm diameter and 10 cm in depth were obtained from N0 and N100 or N140 treatment plots in Fields B and C and from each treatment block in Field A during the growing season for the measurement of soil bulk density and gravimetric water content. Soil cores were dried at 105°C until constant weight, and dry weights were used for determining soil gravimetric water content. Soil water content was expressed as volumetric soil water content and as percentage WFPS. Equation [2] was used to calculate percentage WFPS:

$$\% \text{WFPS} = \left| \frac{\text{MC} \times P_{\text{B}}}{\left( 1 - \frac{P_{\text{B}}}{P_{\text{P}}} \right)} \right| *100$$
[2]

where MC is the gravimetric soil water content (g g<sup>-1</sup>),  $P_{\rm B}$  is the soil bulk density (Mg m<sup>-3</sup>), and  $P_{\rm p}$  is the soil particle density (2.65 Mg m<sup>-3</sup>) (Linn and Doran, 1984).

## Laboratory Soil Incubation Experiment

#### Soils

Six to seven intact soil cores were collected from each treatment plot between rice plants during the week before drainage when the field was still flooded. Undisturbed soil samples were collected using a flexiglass tube (12-cm length, 4.6-cm i.d.). Floodwater (2 cm) was included as part of the sample core to maintain the anoxic soil conditions. Immediately after soil sampling, the entire tube was sealed at both ends and stored at 5°C before the laboratory experiment.

#### Laboratory Experimental Setup

The flooded soil cores were placed in a flow-through incubator and allowed to dry at 21°C with the top end exposed to ambient air. This temperature was chosen as previous field studies showed that mean daily air temperature during final drainage ranged from 19 to 22°C (Adviento-Borbe et al., 2013; Pittelkow et al., 2013). The incubation period ended when the soil had dried out as indicated by no weight reduction from one sampling period to the next and gas flux rates were near zero. The endpoint of the incubation varied among soils because different soil textures required variable drying times (89, 42, and 50 d for Fields A, B, and C, respectively). For each treatment, gas flux measurements were replicated six times. To account for ambient CH<sub>4</sub> and N<sub>2</sub>O concentrations during incubation period, three 2-L glass jars with empty flexiglass tube were included and labeled as controls. After the incubation, ambient gas concentrations inside these jars were 0.318 to 0.414 mg  $N_2O L^{-1}$ , 1.55 to 2.23 mg  $CH_4 L^{-1}$ , 0  $\mu$ g N<sub>2</sub>O-N core<sup>-1</sup> d<sup>-1</sup>, and 39.7  $\mu$ g CH<sub>4</sub>-C core<sup>-1</sup> d<sup>-1</sup>.

#### Laboratory Gas Sampling and Analyses

Daily gas sampling was performed on soil cores for 3 wk immediately after the disappearance of floodwater but when the soils were still water saturated. Before and after this 3-wk intensive sampling, gas sampling occurred once or twice a week. For each sampling event, a soil core was placed inside a 2-L glass jar with a screw-cap lid in which a septum was fitted for gas flux determination. Jars were closed for 30 min during gas sampling and a 25-mL headspace gas sample was taken at 0, 10, 20, and 30 min after jar closure and was immediately transferred into evacuated 12-mL glass vial (Labco Ltd.) with rubber septa double sealed with 100% silicon for leak-free storage before gas analysis. The procedure to analyze N2O and CH4 concentrations and calculations of GHG emissions followed the method used in the field measurements as described above. To determine if contamination in the incubation setup occurred, the emissions of CH<sub>4</sub> and N<sub>2</sub>O from the soil cores were compared with the amount of CH<sub>4</sub> and N<sub>2</sub>O emitted from the control jars. Throughout flux measurements, GHG emissions from the jars at time zero were close or equal to zero (data not shown) based on significance test at P value < 0.05. Final CH<sub>4</sub> and N<sub>2</sub>O flux rates were expressed as micrograms C or N per core per day.

#### Calculation and Expression of Soil Water Content

After each gas-sampling event was completed, the heights of the soil core, standing water (during flooded), headspace from surface of soil core to tip of flexiglass tube (during drained or dried out), and weight of the entire soil core were determined. These data were used to calculate soil bulk density, soil water content, and GHG flux estimates. The dry weight of each soil core was obtained at the end of the incubation period by drying all soil cores at  $105^{\circ}$ C until constant weight. Soil bulk density and porosity ranged from 0.92 to 1.31 Mg m<sup>-3</sup> and 50.5 to 71.4%, respectively, depending on soil and time during incubation.

#### Soil Nitrogen Content

Inorganic N (NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>) in the 10-cm soil core was quantified on a 10 g (dry wt. equivalent) subsample by destructive sampling of cores. Soil N contents were measured from all the treatments four to five times (three replicates per sampling time) during the entire incubation period. Well-mixed fresh soil was added to 100 mL of 0.5 M K<sub>2</sub>SO<sub>4</sub> and an hour of shaking followed by filtration through Whatman no. 42 filter paper (Whatman Ltd.). Soil (NO<sub>3</sub> + NO<sub>2</sub>)-N was determined using the vanadium (III) chloride reduction method (Doane and Horwáth, 2003) and the salicylate method for soil (NH<sub>4</sub> + NH<sub>3</sub>)-N (Verdouw et al., 1978; Forster, 1995). Soil inorganic NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> contents were expressed as milligrams N per core.

#### **Data Analysis**

All data were subjected to normality tests using the Shapiro–Wilk approach (P = 0.46 to <0.001) and gas flux data that failed the test were analyzed using one-way nonparametric procedure (SAS Institute, 2010). For the laboratory soil incubation study, differences in inorganic soil N contents among treatments and soil types were analyzed using PROC MIXED with least significant difference tests at P value < 0.05, while differences in GHG flux rates among N treatments (Fields B and C) and fields were analyzed using the Wilcoxon scores (SAS Institute, 2010). Within each field, dependence of measured CH<sub>4</sub> and N<sub>2</sub>O gas flux rates on soil water content was described with a nonlinear regression model (Gauss–Newton equation):

$$F = a * e^{\left[-5*\left(\frac{\text{WFPS}-b}{c}\right)^2\right]}$$

[3]

where F is the daily  $CH_4$  or  $N_2O$  flux rate (µg core<sup>-1</sup> d<sup>-1</sup>); and a, b, and c coefficients denote the maximum  $N_2O$  or  $CH_4$ flux rate, soil water content at maximum gas flux rate, and the size of the range of soil water content with significant gas flux rates, respectively (SYSTAT Software, 2008). A t-test was applied to examine statistical significance of the parameter estimates generated by the Gauss-Newton model (SYSTAT Software, 2008). Differences in field fluxes of CH<sub>4</sub> and N<sub>2</sub>O among treatments within each field during drained period were analyzed using PROC MIXED with least significant difference tests at P < 0.05, while analysis of repeated measures were conducted using AR1 (autocorrelation 1) for differences of GHG fluxes due to date of measurement (SAS Institute, 2010). Relationships between measured dissolved  $CH_4$ , soil exchangeable N content, and GHG emissions in the laboratory incubation and field studies were analyzed using the Pearson correlation. To calculate the proportion of drainage emissions with the corresponding percentage WFPS to total GHG emission we used linear interpolation.

# **Results and Discussion**

# Field Experiment: Dissolved Methane and Greenhouse Gas Emissions Late in the Growing Season

Across all fields, dissolved CH<sub>4</sub> concentrations in soil pore water ranged from 0.12 to 15.6  $\mu$ g CH<sub>4</sub> mL<sup>-1</sup> during the last month of flooding before the final draining event and became negligible after the final drainage (Fig. 1). Dissolved CH<sub>4</sub> differed among the three fields with the mean daily dissolved CH<sub>4</sub> concentrations declining in the order of Field B > Field A > Field C (Fig. 1). During this period, dissolved CH<sub>4</sub> concentrations were not significantly different in fields receiving different N fertilizer rates (Fields B and C; data not shown) (P = 0.158– 0.194). In Field A, the large flux of dissolved CH<sub>4</sub> following drainage is presumably caused by contamination of CH<sub>4</sub> in the gas phase as it flows into the cracks around the pore sampler during soil solution sampling (Fig. 1). Soil cracking around the sampler during soil drying was observed because of the relatively large diameter of pore sampler used in Field A.

Daily  $CH_4$  fluxes were 282 to 2153 g  $CH_4$ -C ha<sup>-1</sup> d<sup>-1</sup> at all locations during the last month of flooding before final drainage (Fig. 1). Fluxes were similar in all N-rate treatments during this submerged period, indicating that higher N fertilizer rates in Fields B and C had little to no effect on CH<sub>4</sub> emission during this period. Other studies have also reported that fertilizer N rates have a minimal effect on CH<sub>4</sub> emissions in flooded rice systems (Schütz et al., 1989; Xie et al., 2010; Cicerone et al., 1992; Linquist et al., 2012a; Pittelkow et al., 2014). Daily CH<sub>4</sub> fluxes at all locations after drainage were up to 1.6- to 2.7-fold larger than what was emitted before drainage and high rates of CH<sub>4</sub> emissions occurred within 1 to 3 d after the floodwater disappeared (Fig. 1). During this dry-down period, soil WFPS at 0 to 10 cm of soil depth ranged from 87 to 51% (36–55% soil volumetric water content,  $MC_v$ ), 132 to 49% (61 to 23%  $MC_v$ ), and 125 to 55% (76 to 34%  $MC_v$ ) for Fields A, B, and C, respectively. The largest CH<sub>4</sub> emissions occurred at 72 to 94% WFPS in all fields while largest N2O emissions occurred only in Field C at 63 to 85% WFPS. On



Fig. 1. Dissolved  $CH_4$  and greenhouse gas emissions in the three fields at heading to harvest during the 2011 rice growing season. Shaded area indicates flooded period. Standard errors were based on three replicate blocks. M206 is rice variety used in Field A and N0, N100, N140, N200, N260 in Fields B and C indicate N rates of 0, 100, 140, 200, and 260 kg urea N ha<sup>-1</sup>, respectively.

average, 10% (5 to 14%) of the total seasonal  $CH_4$  emissions occurred across all fields during postdrainage (Pittelkow et al., 2013; Adviento-Borbe et al., 2013; Simmonds et al., 2015). These results are in agreement with other field studies (Watanabe et al., 1994; Bossio et al., 1999; Denier van der Gon et al., 1996; Yagi et al., 1996).

Postdrainage CH<sub>4</sub> emissions were significantly related to the amount of predrainage dissolved  $CH_4$  in soil pore water (r =0.482, P = 0.001) in these fields. This is also seen by the linear relationship between dissolved CH<sub>4</sub> in soil pore water just before the drain and cumulative CH4 emissions after the drain event (Fig. 2). In flooded rice systems, Lindau (1994) and Bossio et al. (1999) also found a strong relationship between dissolved CH and CH<sub>2</sub> emissions at the end of the season. When soils started to dry, the concentrations of dissolved CH<sub>4</sub> in the soil water pore were no longer proportional to the CH<sub>4</sub> emission rates that were released either through the rice plants or soil surface (Fig. 1). In fact, maximum rates of  $CH_4$  emission occurred at the very low levels of dissolved  $CH_4$  (<0.3 µg  $CH_4$  mL<sup>-1</sup>) at the first few days of drainage. These results suggest that the rapid release of large amounts of CH4 during soil draining is likely caused by gasification of dissolved CH<sub>4</sub> in the soil pore water and the escape of a large CH<sub>4</sub> pool accumulated in the gas phase as has been reported by others (Cicerone et al., 1992; Denier van der Gon et al., 1996).

Since  $CH_4$  emissions were measured from the system where both the soil and rice plant were enclosed, we cannot identify the proportion of postdrainage  $CH_4$  flux being released through the rice plant vs. from the soil surface. Based on an isotope study, Han et al. (2005) reported that the sharp increase in  $CH_4$  emission following drainage resulted from enhanced transport from the soil surface.

Nitrous oxide emissions were not observed in any field before drainage because of anaerobic soil conditions (Fig. 1), as expected from continuously flooded rice fields (Smith et al., 1982; Zou et al., 2005). However, a large portion (33–79%) of the total growing season N<sub>2</sub>O emissions occurred following drainage when soils dried out, creating ideal conditions for nitrification and denitrification to co-occur (Ghosh et al., 2003; Knowles, 2005). Like  $CH_4$ , N<sub>2</sub>O emissions were variable across all fields after the final drainage. There were no consistent effects of N fertilizer rate on N<sub>2</sub>O emissions; however, daily



Fig. 2. Relationship of dissolved  $CH_4$  in soil water pore just before the drain period and cumulative  $CH_4$  emissions after the drain. Data were from all treatments and fields.

 $N_2O$  emissions were significantly higher at the highest N rate (N260) (57 gN<sub>2</sub>O-N ha<sup>-1</sup> d<sup>-1</sup>) (P < 0.0001) in Field C, but this was not the case in Field B (Fig. 1). While N<sub>2</sub>O emissions have been shown to be related to N inputs in rice systems, higher emissions are more typically associated with excess N rates (Pittelkow et al., 2014). In this study, all of our measurements were taken at the end of the growing season when mineral N is expected to be low (Linquist et al., 2006). In Field C, it is possible that at the high N rate (N260), all fertilizer N may not have been taken up and excess fertilizer N in the soil may have contributed to N<sub>2</sub>O emissions following drainage.

Considering the total postdrainage emissions,  $N_2O$  emissions accounted for 0 to 82% (mean 27%) of the total growing-season  $N_2O$  emissions (Adviento-Borbe et al., 2013; Pittelkow et al., 2013; Simmonds et al., 2015), which is in accordance with Zou et al. (2005), who reported that 39% of the total seasonal  $N_2O$  emissions were emitted following final drainage at harvest.

## Laboratory Experiment: Nitrous Oxide and Methane Flux Rates and Soil Water-Filled Porosity

All soils from the three fields exhibited different soil water content profiles during the drying period (Fig. 3). Soil from Field B dried faster than soils from Fields A and C, which had higher clay contents (Table 1). At the start of the incubation, floodwater disappeared after approximately 10 d of drying. Throughout the incubation period, the percentage WFPS ranged from 16 to 159% (6–85%  $MC_v$ ) for Field A, 17 to 160% (10–91%  $MC_v$ ) for Field B, and from 12 to 143% (6–91%  $MC_v$ ) for Field C. Having percentage WFPS of greater than 100% is not uncommon in flooded clayey rice soils. For example, Aulakh et al. (1996) reported higher than 100% and even as high as 164%. In our case, the reason for this is that these soils have significant amounts of montmorillonite and other 2:1 clays (Table 1) that swell on wetting and thus, the particle size density changes (Haines, 1923; Nadeau, 1985; Ringrose-Voase et al., 2000), which is not accounted for in our estimation of percentage WFPS from Eq. [2].

Similar to field measurements,  $CH_4$  fluxes were near zero before the drainage (i.e., at the start of the incubation when soils from the three fields were flooded) and when these soils had dried out. During soil drying,  $CH_4$  fluxes were detected at similar WFPS for the different soils: 67 to 156% WFPS (47–74% MC<sub>v</sub>) for Field A, 51 to 139% WFPS (28–63% MC<sub>v</sub>) for Field B, and 47 to 113% WFPS (33–68% MC<sub>v</sub>) for Field C (Fig. 4d–f). Almost 95% of the detected postdrainage  $CH_4$  emissions occurred between 100 and 76% WFPS. Maximum daily  $CH_4$  fluxes were similar across fields and occurred on average at 88% WFPS. Similarly, Hou et al. (2012) reported  $CH_4$  peak emissions occurring at 99% WFPS and emissions declining as WFPS decreased to 84%.

For soils from Fields B and C, which had received different N fertilizer rates, there was no effect of N treatment on  $CH_4$  fluxes, similar to findings observed in the field (Fig. 1). Throughout the incubation period,  $CH_4$  was not correlated with soil  $NH_4^+$  content (P = 0.166-0.760), and  $NH_4^+$  concentrations were always low to medium range (0.4–20 mg kg<sup>-1</sup>) (Fig. 5). Low  $NH_4^+$  values at the end of the growing season are expected as fertilizer N and indigenous soil N is



Fig. 3. Changes in percentage soil water-filled pore space during soil drying in the three fields. Values were based on duplicate measurements and standard errors were excluded in the plot for clarity of soil water content profile during incubation.

taken up by the crop during the season (Linquist et al., 2006). While  $\mathrm{NH}_4^+$  has been shown to affect  $\mathrm{CH}_4$  production and emissions (Schimel, 2000), the  $\mathrm{NH}_4^+$  levels in this study may have been too low to have an effect.

Nitrous oxide fluxes varied across fields and the highest emissions occurred when soils were drier than when the maximum  $CH_4$  fluxes were observed (Fig. 4a-c). Fluxes of  $N_2O$  occurred between 45 and 109% WFPS (35–52% MC<sub>y</sub>) for Field A, 24 to 104% WFPS  $(23-53\% MC_v)$  for Field B, and 16 to 71% WFPS (16-37% MC<sub>v</sub>) for Field C. When daily N<sub>2</sub>O fluxes were fitted against percentage WFPS, the empirical relationships showed that the highest N<sub>2</sub>O fluxes occurred at about 71% WFPS for Fields A and B and at 45% WFPS for Field C (Fig. 4a-c). Our results for Fields A and B are similar to the findings of Schmidt and Kaupenjohann (2000), where N<sub>2</sub>O peaks occurred at 74% WFPS for arable soils. However, Zheng et al. (2000) and Hou et al. (2012) reported that large amounts of N<sub>2</sub>O emissions occurred at higher water contents (82-108% WFPS). In this study, approximately 95% of measured postdrainage N<sub>2</sub>O emissions occurred between 43 and 78% WFPS.

While N<sub>2</sub>O fluxes were observed in all incubation study cores, N<sub>2</sub>O fluxes were not observed in all field studies (Fig. 1). Similar to field flux measurements, the N<sub>2</sub>O flux rates for the soil cores were highest in Field C. The low fluxes in laboratory incubations suggested that we may not have been able to detect these emissions in the field studies because of the size of chambers being used (chambers at the end of the season are 1 m in height to accommodate the rice plants inside). For soils that received different N fertilizer rates (Fields B and C) during the early growing season, mean N<sub>2</sub>O flux rates were not affected by N rates (data not shown) (P = 0.116). In fact, there was no relationship between soil NH<sub>4</sub><sup>+</sup> or NO<sub>3</sub><sup>-</sup> content and soil N<sub>2</sub>O fluxes during soil incubation (P = 0.108-0.582) (Fig. 5).

Although flux measurements from the laboratory soil cores do not fully represent the field emissions, the GHG emission dynamics were similar. In both studies (field and laboratory), we observed that  $CH_4$  was emitted first and was followed by  $N_2O$  as soils dried. Also, in both studies, emissions of  $N_2O$  were higher



Fig. 4. Relationship of CH<sub>4</sub> and N<sub>2</sub>O flux rates and percentage water-filled pore space in the three rice soils incubated at 21°C. Note differences in scale for N<sub>2</sub>O fluxes. Plots (a), (b), and (c), correspond to soil N<sub>2</sub>O flux rate profiles in Fields A, B, and C, respectively, and plots (d), (e), and (f) correspond to soil CH<sub>4</sub> flux rate profiles in Fields A, B, and C, respectively.

in Field C. These results suggest that similar controlling factors influencing gas release occurred in both field and laboratory studies. Simulating field experiment through soil incubation and measurement of GHG fluxes from a vented container has been the approach used by many researchers to further investigate the major factors controlling GHG emissions from rice fields (i.e., Wang et al., 1993; Zheng et al., 2000; Han et al., 2005). Furthermore, since the measurement of GHG emissions from the field did not allow separation of net GHG emissions into soil and plant-mediated emissions, it was still not sufficient to conclude that the bulk of GHG emissions during drainage came from the soil surface. Although sources of GHG fluxes can be measured, particularly during postdrainage using flux chambers with and without rice plants inside installed in the fields, this application may have significant chamber effects on gas fluxes such as increased in temperature, evapotranspiration, and different drying periods. More studies are needed to verify the proportion of total GHG emissions from the soil surface and from the rice plant system.

Overall, our results show that high postdrainage  $CH_4$  emissions occurred at higher percentage WFPS (76–100% WFPS) than for N<sub>2</sub>O (43–78% WFPS), suggesting that a frequent and extended gas sampling scheme is necessary

to capture peaks of both  $CH_4$  and  $N_2O$  emissions. The percentage WFPS quantitatively describes the absolute available water for microbial activities and pathways for trace gases and substrates across various soil textures (Linn and Doran, 1984); however, percentage WFPS varies with soil texture and may not be practical to determine under field conditions since it entails soil sampling, weighing, and oven-drying to determine bulk density and volumetric water content that extend more than a day. From a practical point of view, daily measurements following drainage were required in these rice systems to capture the postdrainage emissions. It required less time to capture postdrainage  $CH_4$  emissions from the field than  $N_2O$  emissions.

#### Conclusions

Reliable estimates of total  $CH_4$  and  $N_2O$  emissions require more accurate and complete accounting of sources of GHG emissions. In flooded rice systems, emissions following final drainage contributed between 5 and 14% and 0 to 82% of the total seasonal  $CH_4$  and  $N_2O$ , respectively. The total amount of  $CH_4$  emitted during the drained period was dependent on the amount of dissolved  $CH_4$  in the soil water before drain. The  $CH_4$  and  $N_2O$  emissions occurred at different percentage



Fig. 5. Soil exchangeable N contents and fluxes of CH<sub>4</sub> and N<sub>2</sub>O in the three fields during selected periods of soil drying. Soil NH<sub>4</sub>–N and NO<sub>3</sub>–N contents followed by the same letter or Roman numeral are not significantly different at P < 0.05. Standard errors were calculated from three replicate blocks.

WFPS values. Although percentage WFPS is an effective indicator of soil water content, it may not be suitable for field measurements. Thus, frequent gas sampling is required to capture peak  $CH_4$  and  $N_2O$  emissions following drainage. Given the relatively large contribution of drainage GHG emissions, we suggest that GHG emissions during drainage be systematically included in the sampling protocols of total GHG emissions to reduce uncertainty and improve the integration of field measurements to larger scales.

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