Soil gas fluxes of N_2O, CO_2 and CH_4 under elevated carbon dioxide under wheat in northern China

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Abstract
Fluxes of nitrous oxide (N_2O), carbon dioxide (CO_2) and methane (CH_4) were measured from soils under ambient (420 ± 18 µmol/mol) and elevated (565 ± 37 µmol/mol) [CO_2] at the Free-Air Carbon dioxide Enrichment (FACE) experiment in a wheat field in northern China. N_2O and CO_2 emissions under elevated CO_2 were increased by 47% ($p < 0.05$) and 11% ($p = 0.063$), respectively, but had no effect on CH_4 flux. A significantly greater emission of N_2O (1812%) and CO_2 (69%) was observed from high-N (190 kg N/ha) than low-N (50 kg N/ha) plots only after simultaneous addition of water and urea. The fluxes of N_2O and CO_2 were positively and significantly correlated with both soil moisture and organic C contents, but CH_4 flux with organic C content only. There was no significant relationship between soil mineral N and gas fluxes.

Key Words
Free air carbon dioxide enrichment, nitrous oxide, carbon dioxide, methane, nitrogen fertilizer, wheat soil.

Introduction
Nitrous oxide (N_2O), carbon dioxide (CO_2) and methane (CH_4) are important greenhouse gases to agriculture. Elevated [CO_2] affects crop growth resulting in changes in soil C and N dynamics as well as greenhouse gas fluxes (Liu and Greaver 2009). Yet, the information on the responses to elevated [CO_2] of these gas fluxes from cropping systems is limited. Emissions of N_2O have been reported to either increase with soil moisture and C input under elevated [CO_2], or decrease as a result of N immobilization (Hungate *et al.* 1997; Arnone and Bohlen 1998). CO_2 emissions were mostly stimulated under elevated [CO_2], which was attributed to enhanced microbial and root respiration (Zak *et al.* 2000; Kou *et al.* 2007). The enhanced CH_4 emissions under elevated [CO_2] in rice paddies were associated with greater amount of root exudates, root autolysis products and tillers (Inubushi *et al.* 2003), but the response of CH_4 oxidation in dryland cropping systems remains unclear. Application of N-fertilizer is a major contributor to greenhouse gas emissions. A recent review indicated that N stimulation of global N_2O and CH_4 emission from multiple ecosystems, including croplands, could largely offset (53–76%) the reduction of CO_2 (via global terrestrial C sink) (Liu and Greaver 2009). However, the interaction between N addition and elevated [CO_2] needs further research. The present study was conducted on a wheat field in northern China using Free-Air Carbon dioxide Enrichment (FACE) facility. The objectives are to investigate (i) the interactive effect between elevated [CO_2] and fertilizer-N application on the fluxes of N_2O, CO_2 and CH_4; and (ii) the relation between these fluxes and soil moisture, organic C and mineral N contents.

Methods
Experimental site
The study site is located in an experimental farm in a soybean-wheat rotation on meadow cinnamon soil in Changping, Beijing, China (40°10'N, 116°14'E), with an average rainfall and temperature of 165 mm and 7.9°C during wheat growing season. The present experiment was conducted from early May to late June in 2008 (from booting to harvest of wheat crop).

Carbon dioxide elevation
The elevation of [CO_2] was achieved from FACE system, consisting of 12 4 m diameter experimental areas, six elevated and six ambient. The two target CO_2 concentrations were 420 (ambient) and 565 µmol/mol (elevated). Carbon dioxide exposure commenced at sowing time and terminated at harvest time.

Wheat cultivation, fertilization and irrigation
Winter wheat (*Triticum aestivum* L. cv. Zhongmai 175) was sown on 10 October 2008 with a seeding rate of

150 kg/ha. The experimental site was fertilized with granular urea before sowing and at jointing stage at a total rate of 50 and 190 kg N/ha to low-N (LN) and high-N (HN) plots, respectively. The immediate (1 h) and short-term (4 h) effects of irrigation on gas fluxes were tested by adding 1.5 L water (equivalent to 75 mm rainfall) to microplots in each treatment at 0900 h on 7 June. The granular urea dissolved in 0.5 L water was applied to HN microplots at 95 kg N/ha and same amount water without urea was applied to LN microplots on 10 June, and gas samples were collected one hour later.

Gas sampling and flux determination

Gas samples for N_2O, CO_2 and CH_4 analysis were taken from closed flux chamber (0.15 m height by 0.16 m diameter) on 5, 15 and 25 May and 4, 7 and10 June between 1300 and 1500 h of the day, and one additional measurement was taken between 1000 and 1200 on 7 June one hour after irrigation. One chamber was inserted a day before the first measurement to a soil depth of 70 mm, and remained *in situ* throughout the experimental period. On each sampling day, the chamber was closed for 0.5 h prior to the first gas sampling. Three gas samples were then collected from the chambers at 10 minute intervals (chambers remained closed) using a gas tight syringe through a rubber bung. Gas of 30 mL was transferred into evacuated glass vials and transported to the laboratory within the same day for analysis by gas chromatography. Flux rates of N_2O, CO_2 and CH_4 were calculated from the linear change in gas concentrations in the chamber.

Soil analysis

Two auger samples (0–0.1 m) of top-soil from each microplot in each experimental ring were bulked. Subsamples (10 g) of the fresh soil were extracted with 2 M KCl in a 1:5 ratio of soil to extractant. The concentrations of ammonium (NH_4^+) and nitrate (NO_3^-) in the filtered extract were determined by continuous flow analysis (FOSS Fiastar 5000). Organic C was determined by Walkley-Black partial oxidation method.

Statistical analysis

Data were analysed with MINITAB 14 statistical package using a factorial model analysis of variance with main effects as [CO_2], N application and sampling time. Gas flux rates were regressed against soil properties.

Results

N_2O flux

Positive N_2O fluxes were observed for all treatments over the experimental period. N_2O emissions were 47% higher ($p < 0.05$) from elevated than from ambient [CO_2] plots (Table 1), regardless of N application rate and sampling time. Interaction between N application rate and sampling time was significant ($p < 0.001$), with significantly higher (1812%) flux recorded from HN than LN plots after addition of dissolved urea in 0.5 L water on 10 June, but there were no significant differences at other sampling times (Figure 1a). The addition of 1.5 L water resulted in 37% and 43% increase, though not significant, in N_2O flux from HN plots 1 h and 4 h after irrigation on 7 June (Figure 1a). N_2O fluxes were positively correlated with CO_2 fluxes ($r = 0.87$, $p < 0.001$), soil moisture content ($r = 0.65$, $p < 0.001$) and organic C content ($r = 0.36$, $p < 0.001$).

Table 1. Effect of [CO_2] on N_2O, CO_2 and CH_4 fluxes (mean ± SE, *n* = 42).

[CO_2] (μmol/mol)	N_2O flux (μg N_2O-N/m^2/h)		CO_2 flux (mg CO_2-C/m^2/h)		CH_4 flux (μg CH_4-C/m^2/h)	
420	25.6 ± 5.7	*	39.7 ± 4.4	†	–5.5 ± 1.0	ns
565	37.6 ± 9.6		44.0 ± 4.9		–4.6 ± 0.8	

ns: not significant; † $p < 0.1$; * $p < 0.05$

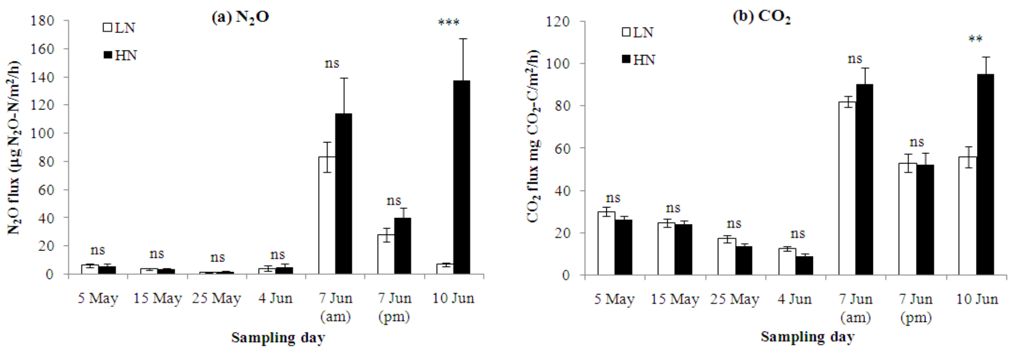

Figure 1. The effect of N addition and irrigation on the fluxes of (a) N_2O and (b) CO_2 in LN and HN plots. Bars indicate standard errors. ns, no significant difference, ** $p < 0.01$, *** $p < 0.001$.

1 – 6 August 2010, Brisbane, Australia. Published on DVD.

CO_2 flux
CO_2 fluxes during the experimental period were always positive. Elevated [CO_2] marginally increased (p = 0.063) CO_2 emission by 11% (Table 1), regardless of N treatment and sampling time. Interaction between N application rate and sampling time was significant ($p < 0.001$), with 69% increase in CO_2 emission from HN compared to LN plots on 10 June, but no significant differences were seen during other sampling times (Figure 1b), which was consistent with the aforesaid significant increase in N_2O flux. CO_2 fluxes were positively correlated with soil moisture ($r = 0.87, p < 0.001$) and organic C contents ($r = 0.30, p < 0.01$).

CH_4 flux
Net CH_4 consumption was detected at all samplings, except on 10 June from HN plots with 1.65 and 0.12 μg CH_4-C/m^2/h under ambient and elevated [CO_2], respectively. [CO_2] and N application rate had no significant effect on CH_4 fluxes throughout the course of the measurement period. CH_4 fluxes were positively correlated with organic C content ($r = 0.32, p < 0.01$).

Soil moisture, mineral N and organic C
Gravimetric soil moisture content averaged 4–9% in the early stage of experimental period and increased ($p < 0.001$) to 15–18% after water addition on 7 and 10 June, but no significant effect of [CO_2]. Elevated [CO_2] increased organic C by 11% ($p < 0.01$) in HN plots, but not in LN counterparts. More NH_4-N (27%, $p < 0.05$) and NO_3-N (250%, $p < 0.001$) was detected from HN than LN plots, regardless of [CO_2] and sampling time. Soil mineral N did not significantly affect the fluxes of the three gases.

Discussion
Effect of [CO_2], N addition and irrigation on soil N_2O emissions
Elevated [CO_2] significantly increased N_2O emissions by 47%. This is in agreement with a 40% increase in N_2O emissions recorded in a mixed perennial ryegrass/white clover sward in a FACE experiment (Baggs *et al*. 2003), and a 30% (though not significant) increase in a glasshouse experiment growing timothy under elevated [CO_2] (Kettunen *et al*. 2005). The increase was possibly due to a stimulation of root biomass and root exudation for denitrifiers (Arnone and Bohlen 1998; Kettunen *et al.* 2007), as suggested by the positive correlation between N_2O and CO_2 fluxes, as well as the significant increase in organic C content under elevated [CO_2] in HN plots. Nitrogen addition increased N_2O flux throughout the experimental period, with significant increase after irrigation and additional N treatment, for both CO_2 concentrations. This is expected as nitrification is mainly stimulated by the NH_4-N (Bouwman *et al.* 1993) while denitrification is enhanced by high soil moisture and NO_3-N contents (Bolan *et al.* 2004). The interaction between elevated [CO_2] and N application rate on N_2O flux was not significant. This finding differs from Baggs *et al*. (2003) who showed a significant increase in N_2O emissions only under 560, but not 140 kg N/ha/y. This implies at low N application rates, N supply might be a limiting factor for N_2O emissions despite the greater C availability under elevated [CO_2]. This is possible as N immobilization was observed as a result of enhanced root production and the higher C/N ratio of residue under elevated [CO_2] (Hungate *et al.* 1997; Pleijel *et al.* 1998). In the present study, elevated [CO_2] increased N_2O emissions under both LN and HN plots, which suggests that N is not limiting. This highlights the importance of striking a balance between the increases in N_2O emissions and the grain yield achieved by applying N-fertilizer.

Effect of [CO_2], N addition and irrigation on soil CO_2 emissions
Elevated [CO_2] marginally increased soil CO_2 efflux, which is consistent with results obtained from other FACE sites growing cotton (Wood *et al*. 1994), spring wheat (Prior *et al*. 1997), winter wheat (Kou *et al*. 2007) and soybean (Peralta and Wander 2008). The stimulation of efflux was associated with increased biomass under elevated [CO_2] (Kimball *et al.* 2002). Increases in CO_2 emission were observed however only when both water and urea were simultaneously added, indicating they were important factors affecting the immediate response of soil microbes. In summary, the negative environmental impact of elevated CO_2 on greenhouse gas emissions from soil need to be balanced by reductions in CO_2 resulting from increased C capture in crop biomass.

Effect of [CO_2] and N addition on net CH_4 consumption
During most of the study period, there was a net oxidation in CH_4, which is common for agricultural soils in temperate and well-drained sites (Ineson *et al*. 1998). Net CH_4 consumption was overall lower, albeit not significantly, under elevated than ambient [CO_2] (Table 1). This is consistent with Ineson *et al*. (1998), who observed an average net CH_4 consumption of 25.5 and 8.5 μg/m^2/h respectively for the control and enhanced

CO_2 plot of perennial ryegrass. Incubation of soils taken from forest also indicated a significantly lower net CH_4 consumption under elevated than ambient [CO_2] (Philips *et al.* 2001; McLain and Ahmann 2008). These results indicate that less CH_4 being removed from the atmosphere under elevated [CO_2], which is comparable to a higher CH_4 production under elevated [CO_2] in wetland systems like rice paddies (Inubushi *et al*. 2003). Increases in CH_4 production (or reduction in CH_4 oxidation) under elevated [CO_2] are associated with the stimulation of methanogenic activity (Wang and Adachi 1999), reduction of methanotropic activity (Philips *et al.* 2001) and/or increased moisture in deep (>25 cm) soils (McLain and Ahmann 2008). The effect of N addition was not significant in the present study, which agrees with Ineson *et al*. (1998). However, an interaction between [CO_2] and N addition on CH_4 oxidation was observed by Baggs and Blum (2004).

Conclusions

Fluxes of N_2O and CO_2 from the soil were positive while CH_4 negative overall under elevated [CO_2]. Under elevated [CO_2], the higher N_2O and CO_2 emissions probably resulted from increases in substrate availability while net CH_4 consumption was not changed. N addition generally increased N_2O and CO_2 production, particularly when the soil was wet, but not CH_4. There was no interaction between elevated [CO_2] and N addition on the fluxes of these gases in the short-term from this wheat field. These findings have major implications on global climate change. In particular, there is a positive feedback relation between elevated [CO_2] and the concentrations of these gases. Although N application generally boosts crop yield, in terms net greenhouse gas production, excessive N application will offset only part of the extra C sequestered, and should be avoided from both environmental and financial grounds.

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1 – 6 August 2010, Brisbane, Australia. Published on DVD.