Biogeosciences Discuss., 8, 9577–9607, 2011
www.biogeosciences-discuss.net/8/9577/2011/
doi:10.5194/bgd-8-9577-2011

Biogeosciences Discussions

Responses of N_2O and CH_4 fluxes to fertilizer nitrogen addition rates in an irrigated wheat-maize cropping system in northern China

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Received: 22 August 2011 – Accepted: 19 September 2011 – Published: 22 September 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Model and field studies generally posit that when the application rates of nitrogen fertilizer exceed crop needs, nitrous oxide (N_2O) emissions will increase nonlinearly, though linear responses are also extensively reported by field studies. We conducted year-round measurements of crop yield, N_2O and methane (CH_4) fluxes for treatments of six nitrogen levels (0, 135, 270, 430, 650 and 850 kg N ha^{-1} yr^{-1} in the form of urea) in a typical irrigated wheat-maize rotation field in northern China. Linear models characterized the responses of cumulative N_2O emissions to fertilizer rates well; therefore, the calculated N_2O emission factors of 0.17 ± 0.03 %, 0.73 ± 0.07 % and 0.49 ± 0.06 % for the wheat season, maize season and annual scale, respectively, were appropriate for the different fertilizer rates. The cumulative CH_4 uptake by the soil tended to be enhanced at higher fertilizer rates (≥350 kg N ha^{-1}) in the maize season whereas no effect was observed for the wheat season. The crop yields stopped increasing at fertilizer rates greater than 650 kg N ha^{-1} yr^{-1}. When the annual fertilizer rates increased from 270 to 430, from 270 to 650 and from 270 to 850 kg N ha^{-1} yr^{-1}, the crop yields increased only 4–15 % (0.6–2.2 t ha^{-1} yr^{-1}), but cumulative N_2O emissions increased 36–115 % (0.9–3.0 kg N ha^{-1} yr^{-1}). We recommend 270 kg N ha^{-1} yr^{-1} as the locally optimum fertilizer rate. Considering the N inputs by fertilization (270 kg N ha^{-1} yr^{-1}), irrigation (4.3 ± 0.2 kg N ha^{-1} yr^{-1}) and deposition (wet deposition: 30.5 ± 1.5 kg N ha^{-1} yr^{-1}), the slightly positive soil N balance could maintain the current crop yield (>14 t ha^{-1} yr^{-1}) and reduce the present high N_2O emissions (>3.51 kg N ha^{-1} yr^{-1}) of the local farmers' practice (fertilizer rate: >430 kg N ha^{-1} yr^{-1}).

1 Introduction

Methane (CH_4) and nitrous oxide (N_2O) are considered the most potent greenhouse gases after carbon dioxide (CO_2). Their global warming potentials on a 100-yr time

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horizon are 25 and 298 CO_2-equivalents, respectively. N_2O also participates in the destruction of stratospheric ozone (O_3) (Forster et al., 2007). Fertilized upland agricultural soils are the most important anthropogenic N_2O sources and are significant CH_4 sinks (Bouwman et al., 2002; Dutaur and Verchot, 2007). The production of N_2O and the consumption of CH_4 in soils are mainly biological processes, and the activity of these microbes is strongly affected by natural conditions and agricultural management (Snyder et al., 2009).

Agricultural management, such as the addition of nitrogen fertilizer, can significantly affect the fluxes of CH_4 and N_2O between the atmosphere and agricultural soils (Bouwman et al., 2002; Bodelier and Laanbroek, 2004; Aronson and Helliker, 2010). It has been generally accepted that the application of nitrogen fertilizer, especially at high rates (>100 kg N ha^{-1} yr^{-1}), may inhibit CH_4 uptake by upland agricultural soils (Aronson and Helliker, 2010). However, the application of nitrogen fertilizer has also been reported to produce no effects and enhanced effects on CH_4 uptake (Bodelier and Laanbroek, 2004). The factors, such as crop type, agricultural management, precipitation, quantity, type, timing and history of fertilization, need to be accounted for the diverse effects.

The addition of nitrogen fertilizer increases N_2O emissions from cultivated soils. The relationship between N_2O emissions and fertilizer rates is complex. The response curves of N_2O emissions as a function of the fertilizer rate are not common. Only a few studies that have utilized fertilized treatments with more than two nitrogen levels could obtain functional relationships between N_2O emissions and fertilizer rates (Grant et al., 2006; Mosier et al., 2006; Halvorson et al., 2008; Ma et al., 2010; Hoben et al., 2011). When fertilizer rates are in excess of the amount required to optimize crop growth, N_2O emissions are stimulated (McSwiney and Robertson, 2005; Zebarth et al., 2008). Some studies reported the N_2O emissions exponentially increased with fertilizer rates (Grant et al., 2006; Ma et al., 2010; Hoben et al., 2011), but because of the complexity of agricultural management and natural conditions, the functional relationships may not be nonlinear. Hoben et al. (2011) obtained both linear and nonlinear relationships

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for different years and sites with on-farm maize crops in the US Midwest. Using a 5-year field measurement, Mosier et al. (2006) and Halvorson et al. (2008) reported that the N_2O emissions linearly increased with fertilizer rates in several irrigated cropping systems in Colorado (US).

Wheat and maize together accounted for 50.9 % of the total sowing area and 52.6 % of the yield of grain crops in China (China Statistical Yearbook, 2010). The center of wheat and maize productions geographically locates in northern China (Tong et al., 2003), where the one-year winter wheat-summer maize rotation has been extensively adopted. The highly productive double-cropping system relies on the inputs of irrigation water (90–690 mm yr^{-1}, Wang et al., 2008) and chemical fertilizers (550–600 kg N ha^{-1} yr^{-1}, Ju et al., 2009). With a rapid increase in the fertilizer rate, the use efficiencies of chemical fertilizers evaluated as yield per unit chemical fertilizer sharply decrease (Tong et al., 2003; Ju et al., 2009). The overuse of chemical nitrogen fertilizers creates many environmental problems, such as nitrate leaching, water pollution, soil salination, soil acidification and additional N_2O emissions (Zhen et al., 2006; Ju et al., 2009). Until now, the responses of N_2O emissions, CH_4 uptake and crop yield to increased fertilizer rates in the wheat-maize rotation fields have remained unclear. Studying the responses of N_2O emissions, CH_4 uptake and crop yield to fertilizer rates may help to explain the applicability of a set emission factors for N_2O to different fertilizer rates and may help to determine the optimum fertilizer rate for reducing N_2O emissions, enhancing the CH_4 uptake and simultaneously maintaining crop yield. We therefore created treatments with six nitrogen levels in a typical irrigated wheat-maize rotation field in northern China. The aims of the study were to (a) characterize the responses of crop yield, N_2O and CH_4 fluxes to fertilizer rates; (b) quantify the emission factors of N_2O for different fertilizer rates; and (c) determine the relationships between N_2O/CH_4 fluxes and crop yield and recommend the optimum fertilizer rate for the wheat-maize rotation fields.

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2 Materials and methods

2.1 Experimental site

The experimental site (34°55.51′ N, 110°42.59′ E) is situated within the Dong Cun Farm in the Yongji county, Shanxi province, northern China. The annual mean air temperature and total precipitation were 14.7 °C and 559 mm during the period from 2000 to 2010 (National Climatic Data Center, ftp://ftp.ncdc.noaa.gov/pub/data/gsod). The soil of the experimental field is a cinnamon soil (National Soil Survey Office, 1998) with 31.8 ± 0.9 % clay (<0.002 mm), 38.9 ± 0.6 % silt (0.002–0.02 mm) and 29.3 ± 1.4 % sand contents (0.02–2 mm) in the uppermost 10 cm. The soil (0–10 cm) has an organic carbon content of 11.3 ± 0.6 $g\,kg^{-1}$ and a total nitrogen content of 1.12 ± 0.05 $g\,kg^{-1}$. The soil is alkaline (pH 8.7, 0–10 cm) and has a bulk density of 1.17 ± 0.04 $g\,cm^{-3}$ (mean value ± standard error, 0–6 cm). The history of the management of the experimental field is provided in Liu et al. (2011).

Eighteen experimental plots (6 × 6 m each) with three replicates of six fertilizer nitrogen levels (0, 135, 270, 430, 650 and 850 $kg\,N\,ha^{-1}\,yr^{-1}$, hereafter referred to as N0, N135, N270, N430, N650 and N850) were established by a completely randomized design on 16 October 2009. The plots were cultivated with winter wheat (*Triticum aestivum* L.) at 20–23-cm row spacing on 19 October 2009, and with summer maize (*Zea mays* L.) at a density of 7.3 plants m^{-2} on 16 June 2010. The wheat and maize were harvested on 15 June and 15 October 2010, respectively. The residue was cut into pieces of 5–10 cm after harvest and was mechanically ploughed into the soil (0–20 cm) just before seeding. A manually movable sprinkler irrigation system was used to irrigate the crops by underground water. Herbicide was applied once each wheat and maize growing season. Pesticide was applied only once during the maize growing season. Nitrogen fertilizer in the form of urea was applied three times per year at the sowing time (tillage for 20 cm after surface broadcast) and turning-green stage of wheat (soil covering for 0–5 cm after band application) and at the 18- to 19-leaf stage of maize (soil covering for 0–5 cm after band application) (Table 1). In addition, the calcium

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superphosphate (60.0 kg P ha^{-1} yr^{-1}) and potassium sulfate (30.0 kg K ha^{-1} yr^{-1}) were applied at the wheat sowing time for all treatments.

2.2 Measurement of N_2O and CH_4 fluxes

CH_4 and N_2O fluxes were manually measured either daily for seven days after the nitrogen fertilizer applications or two to three times per week during the remaining measuring period from 19 October 2009 to 15 October 2010, using vented static chamber-gas chromatograph measuring system as described in Yao et al. (2009). Prior to seeding, two types of base collar (length × width = 50 × 50 and 55 × 32 cm) made of stainless steel were inserted 20 cm into the soil in the center of each plot. These collars were temporally removed for tillage and seeding operations and remained in position during the remaining investigation period. We designed the chamber of type I with a top, middle and bottom part (each with length × width × height = 50 × 50 × 50 cm) to adapt the chamber height to the varying crop height. The final height of the type I chamber could reach 150 cm. If the maize height surpassed 150 cm, we adopted the type II chamber (length × width × height = 55 × 32 × 20 cm), separated it vertically into two parts and drilled a hole (diameter: 11 cm) in the center of chamber top. The type II chamber allowed the cornstalk to pass through the chamber top and to only cover the maize root. The gap between the type II chamber and the cornstalk was manually patched by sealing a polyethylene membrane (thickness: 0.1 mm) when the chamber was closed. Rubber seals ensured the gas-tightness for the joints of the different chamber parts. Immediately before closing the chamber, ambient air was taken as the sample of deployment time zero. Deployment times were kept to 20–40 min. Five gas samples in total were taken for the concentration analysis of CH_4 and N_2O. The flux calculation was based on the nonlinear fitting approach, as described by Liu et al. (2010). Sampling at 08:00–10:00 a.m. local time, resulted in the lowest deviation (5.3–8.6 %) for the estimation of cumulative emissions and was optimal for intermittent manual flux measurements in the field (Liu et al., 2010). Therefore, the flux measurements at all replicate plots were simultaneously implemented between 08:00 and 10:00 a.m. All the gas samples were analyzed within 10 h after sampling at a laboratory beside the experimental field.

2.3 Auxiliary measurements

In addition to flux measurements, we also measured crop yield, C and N contents of crop straw and grain, air temperature, air pressure, precipitation, groundwater table, soil temperature, soil moisture, soil ammonium (NH_4^+), nitrate (NO_3^-) and dissolved organic carbon (DOC) contents. At harvest, three replicates (0.36 m^2 each for wheat and 2 m^2 each for maize) for each nitrogen level were harvested to measure the crop yield and aboveground biomass by oven drying at 60 °C for two days. The carbon and nitrogen contents of harvested crop straw and grain were measured by the potassium dichromate-volumetric method and the semi-micro Kjeldahl method, respectively. Chamber air and soil temperatures (5 cm) were simultaneously recorded using digital thermometers (JM624, accuracy: ±0.1 °C, Jinming Instrument Co., Ltd., Tianjin, China) while taking air samples. Topsoil moisture (0–6 cm) was measured daily using the oven drying method when the topsoil was frozen in January 2010 or with a portable moisture probe (ML2x, ThetaKit, Delta-T Devices, Cambridge, UK) during the remaining period. The volumetric water content was converted into water-filled pore space (WFPS) using the determined bulk density of 1.17 $g\,cm^{-3}$ and a theoretical particle density of 2.65 $g\,cm^{-3}$. Precipitation, air and soil temperature (5, 15 and 25 cm) for the experimental field were recorded hourly by the nearby meteorological station (Shanghai Changwang Meteorological Technology Co., Ltd., Shanghai, China). Rain was manually collected using a rain gauge. The groundwater table was measured weekly from four replicated polyvinyl chloride tubes (length: 3 m; diameter: 6 cm) that were vertically buried in the soil. The dissolved organic carbon (DOC) and inorganic nitrogen ($NH_4^+ + NO_3^-$) contents of the topsoil (0–10 cm) were measured from four randomly collected soil samples for each nitrogen level, either once per 1–2 days for two

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weeks after the fertilizations or once every seven to fifteen days during the remaining measuring period. The soil was sieved (3-mm mesh), and 12.0 g of soil was then extracted with 100 ml of de-ionized water for the DOC analysis or 2 M KCl-solution for the inorganic nitrogen analysis (shaking for 1 h). The extracts for the DOC analysis were centrifuged and filtrated with polyethersulfone membrane filters. The extracts, irrigation water samples and rain samples were frozen at −18 °C and were later analyzed with an automated DOC, NH_4^+ and NO_3^- analyzer (San^{++} Continuous Flow Analyzer, Skalar Analytical B.V., Netherlands).

2.4 Statistical analysis

The software packages SPSS Statistics Client 19.0 (SPSS China, Beijing, China) and Origin 8.0 (OriginLab Ltd., Guangzhou, China) were used for the statistical data analysis. A general linear model for repeated measures was applied to analyze the significance of the differences in the N_2O and CH_4 fluxes, soil temperature, moisture, DOC, NH_4^+ and NO_3^- contents between treatments. Nonparametric test of two independent samples was used to analyze the differences in the mean crop yield and nitrogen contents of crop straw and grain between treatments. Linear and nonlinear regressions were used to describe the relationships between crop yield, nitrogen contents of crop straw and grain, soil inorganic nitrogen content, soil DOC content, cumulative N_2O emissions, cumulative CH_4 uptake and fertilizer rates. The significance of linear and nonlinear regressions was determined using an F-test.

3 Results

3.1 Environmental parameters

The annual mean air and soil (5 cm) temperatures at the experimental field were 13.8 ± 0.6 and 13.4 ± 0.5 °C (± s.e.), respectively (Fig. 1a). The annual total precipitation was 666.2 mm, which was 19 % higher than the annual mean during the most

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recent 10 yr. The total amount of irrigation was 407.7 mm (Fig. 1b). The groundwater table varied between −0.43 and −1.14 m with a mean of −0.97 ± 0.02 m (Fig. 1b). The total inorganic nitrogen inputs by wet deposition and irrigation were 30.5 ± 1.5 and 4.3 ± 0.2 kg N ha^{-1} yr^{-1}, respectively (Fig. 1c). The seasonal pattern of soil (0–6 cm) moisture was significantly regulated by precipitation and irrigation (Fig. 1d). The soil WFPS of the experimental field ranged from 17.9 to 82.6 %, with an annual mean of 39.4 ± 0.8 %. The differences in seasonal mean soil temperature and WFPS for the treatments of six nitrogen levels were less than 1.2 °C and 0.9 %, respectively (Table 1). We did not observe determinate effects of an increased fertilizer rate on the soil temperature and moisture.

The soil (0–10 cm) inorganic nitrogen contents varied between 0.2 and 42.1 mg N kg^{-1} of soil dry weight (SDW) for N0. The relatively high soil inorganic nitrogen contents (>25 mg N kg^{-1} SDW) were primarily observed within the 18 days after the wheat sowing on 19 October 2009. During the remaining measuring period, the soil inorganic nitrogen contents for N0 were constantly low (<16 mg N kg^{-1} SDW). The fertilizer nitrogen application significantly increased the soil inorganic nitrogen content ($p < 0.05$, Table 1), and the maximum values were obtained within the 5 days after the fertilization on 13 July 2010, which were 150.1, 198.2, 428.0, 579.6 and 590.5 mg N kg^{-1} SDW for N135, N270, N430, N650 and N850, respectively. The annual mean soil inorganic nitrogen contents for N0, N135, N270, N430, N650 and N850 were 8.0 ± 0.9, 23.5 ± 3.2, 33.4 ± 4.5, 56.1 ± 9.1, 74.1 ± 11.0 and 86.1 ± 13.4 mg N kg^{-1} SDW, respectively, and thus linearly increased with the fertilizer rate ($p < 0.01$, Fig. 2a).

The soil (0–10 cm) DOC contents varied between 12.8 and 83.9 mg C kg^{-1} SDW for N0. The relatively high soil DOC contents (> 50 mg C kg^{-1} SDW) for N0 were concentrated between June and August. The minimum DOC contents were similar (20–24 mg C kg^{-1} SDW) whereas the maximum values (161–536 mg C kg^{-1} SDW) sharply increased with the fertilizer rate for N135, N270, N430 and N650. The soil DOC contents for N850 ranged from 14.8 to 494.2 mg C kg^{-1} SDW. The annual mean soil

DOC contents were 42.1 ± 3.7, 53.9 ± 7.2, 58.4 ± 7.4, 66.5 ± 14.1, 73.0 ± 20.9 and 72.1 ± 19.0 mg C kg^{-1} SDW for N0, N135, N270, N430, N650 and N850, respectively, and thus exponentially increased with the fertilizer rate ($p < 0.05$, Fig. 2b).

3.2 Crop yield

The crop yields for N0, N135, N270, N430, N650 and N850 ranged from 6.3 ± 0.1 to 8.2 ± 0.7 t ha^{-1} for wheat and from 4.9 ± 0.7 to 8.9 ± 0.7 t ha^{-1} for maize (Table 1). The maximum crop yields were obtained at the fertilizer rate of 300 kg N ha^{-1} for wheat and 350 kg N ha^{-1} for maize (Table 1 and Fig. 3a). The relationships of crop yield and fertilizer rate could be characterized by exponentially fitting curves ($p < 0.05$, Fig. 3a). The increased fertilizer rate also significantly increased the nitrogen contents of harvested crop straw and grain, especially for the maize straw (Table 1 and Fig. 3b).

3.3 N_2O and CH_4 fluxes

The N_2O fluxes for N0 varied between 1.1 and 68.4 µg N m^{-2} h^{-1}, with an annual mean of 19.9 ± 1.7 µg N m^{-2} h^{-1}. The relatively high fluxes (>40 µg N m^{-2} h^{-1}) for N0 were observed after the irrigation and precipitation between April and September. The maximal N_2O fluxes were obtained within the 9 days after the fertilization on 13 July 2010, which were 424.2, 640.6, 718.3, 954.3 and 1017.8 µg N m^{-2} h^{-1} for N135, N270, N430, N650 and N850, respectively. The extremely high emissions (>200 µg N m^{-2} h^{-1}) lasted for 6, 8, 12, 14 and 14 days and accounted for 21 %, 27 %, 39 %, 40 % and 38 % of the annual emissions for N135, N270, N430, N650 and N850, respectively. With an increase in the fertilizer rate from 0 to 850 kg N ha^{-1} yr^{-1}, the annual mean N_2O emissions from the wheat-maize rotation field sharply increased from 19.9 ± 1.7 to 115.1 ± 22.4 µg N m^{-2} h^{-1} (Table 2).

The CH_4 fluxes fluctuated between −158.4 and 25.1 µg C m^{-2} h^{-1} (Table 2). The weak CH_4 emissions were rarely observed when the groundwater table was in the range from −0.4 to −0.8 m. The period (from 27 July to 4 August 2010) with maximal

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CH_4 uptakes (−75.4– −158.4 µg C m^{-2} h^{-1}) coincided with a period of elevated soil temperatures (27.6–29.0 °C), optimum soil moisture (WFPS: 30.3–46.6 %) and groundwater table (−0.8– −1.0 m).

The nitrogen fertilizer applications significantly enhanced the N_2O emissions, especially the fertilization on 13 July 2010 (Fig. 4a). The annual cumulative N_2O emissions for N0, N135, N270, N430, N650 and N850 were 1.50 ± 0.26, 2.09 ± 0.20, 2.59 ± 0.33, 3.51 ± 0.26, 5.12 ± 0.78 and 5.57 ± 0.68 kg N ha^{-1} yr^{-1}, respectively (Table 2). The relationship between the cumulative N_2O emissions and the fertilizer rate could be well described by linear models for the wheat season, maize season and annual scale ($p < 0.01$, Fig. 5a). The slopes of linearly fitting curves (equal to the N_2O emission factors) were 0.0017 ± 0.0003 (0.17 ± 0.03 %), 0.0073 ± 0.0007 (0.73 ± 0.07 %) and 0.0049 ± 0.0006 (0.49 ± 0.06 %) for the wheat season, maize season and annual scale, respectively.

The annual cumulative CH_4 uptakes for N0, N135, N270, N430, N650 and N850 were −1.49 ± 0.02, −1.59 ± 0.04, −1.56 ± 0.04, −1.51 ± 0.09, −1.69 ± 0.10 and −1.79 ± 0.07 kg C ha^{-1}, respectively (Table 2). The extremely high fertilizer rates (350–450 kg N ha^{-1}) in the maize season tended to increase the cumulative CH_4 uptake as compared with the low fertilizer rates (0–250 kg N ha^{-1}). The increased fertilizer rate (0–400 kg N ha^{-1}) had no detectable effects on the cumulative CH_4 uptake in the wheat season (Table 2 and Fig. 5b).

3.4 Correlations between soil inorganic nitrogen content, soil DOC content, crop yield and cumulative fluxes

The increased soil inorganic nitrogen and DOC contents induced by the increased fertilizer rate linearly and exponentially enhanced the cumulative N_2O emissions in the maize season, respectively ($p < 0.01$, Fig. 6a, b). The effects of the increased soil inorganic nitrogen and DOC contents on the cumulative N_2O emissions in the wheat season were either less intensive or less detectable as compared with the maize season (Fig. 6a, b).

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The exponential stimulation of N_2O emissions in the maize season followed the increase of crop yield induced by the increased fertilizer rate (Fig. 7). The ratios of cumulative N_2O emissions to maize and wheat yields were exponentially correlated with the fertilizer rate (Fig. 8a). Increased fertilizer rates significantly stimulated the nitrogen loss by N_2O emission per unit of crop yield in the wheat-maize rotation field. The ratio of cumulative CH_4 uptake to crop yield was initially increased and then decreased with increased fertilizer rates for the annual scale. In generally, low fertilizer rates ($\leq$ 135 kg N ha^{-1} yr^{-1}) resulted in relatively high cumulative CH_4 uptake per unit of crop yield in the wheat-maize rotation field (Fig. 8b).

4 Discussion

4.1 Responses of cumulative N_2O emissions and CH_4 uptake to fertilizer nitrogen rates

Both the linear and nonlinear responses of cumulative N_2O emissions to fertilizer rates have been observed in field measurements (McSwiney and Robertson, 2005; Mosier et al., 2006; Halvorson et al., 2008; Zebarth et al., 2008; Ma et al., 2010; Hoben et al., 2011). In our study, the responses of N_2O emissions to fertilizer rates could be well described by linear models for the wheat season, maize season and annual scale. Mosier et al. (2006) and Halvorson et al. (2008) also observed that cumulative N_2O emissions increased linearly in response to fertilizer rates in the irrigated maize fields in Colorado, US. However, in nonirrigated maize fields in Michigan, US, and in Ontario and Quebec, Canada, McSwiney and Robertson (2005), Hoben et al. (2011) and Ma et al. (2010) found that cumulative N_2O emissions exponentially increased with fertilizer rates. The field management of irrigation might be a key factor to regulate the responses of N_2O emissions to fertilizer rates.

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N_2O emissions were only a minor loss pathway of the applied fertilizer nitrogen (0.12 and 0.23 % of applied nitrogen in the wheat and maize seasons) in the wheat-maize rotation fields in northern China because of the low available carbon sources (organic matter: 1.0–1.5 %) and soil WFPS for denitrification (Wan et al., 2009; Ju et al., 2009, 2011). The main loss pathways of fertilizer nitrogen were ammonia volatilization (19.4 and 24.7 % of applied nitrogen in the wheat and maize seasons) and nitrate leaching (2.7 and 12.1 % of applied nitrogen in the wheat and maize seasons) below the root zone because of the high soil pH (7.0–8.5), irrigation and concentrated rainfall in summer (Zhao et al., 2006; Ju et al., 2009). In our wheat-maize rotation field, the ammonia volatilization and nitrate leaching should account for more of the fertilizer nitrogen loss due to the higher soil pH (8.7 in average for the soil depth of 0–10 cm), greater amount of precipitation (666.2 mm) and greater amount of irrigation (407.7 mm). Increased fertilizer rates would primarily stimulate the ammonia volatilization and nitrate leaching in the wheat-maize rotation field. Furthermore, the N_2O production was controlled by the competition of soil nitrogen between plants and N_2O-producing microbes. Increased fertilizer rates continuously and significantly enhanced the plant nitrogen uptake. We did not observe the saturated point of soil nitrogen indicated by declining gains in plant nitrogen yield and sharply stimulating microbial N_2O production in the irrigated wheat-maize rotation field. The likely high loss rates of nitrate leaching and ammonia volatilization and the significant plant nitrogen uptake might contribute to the missing exponential stimulation of N_2O emissions for the treatments with high fertilizer rates.

Aronson and Helliker (2010) performed a meta-analysis on the published CH_4 fluxes in the nitrogen amended and control plots. The results indicated that low fertilizer rates (<100 kg N ha^{-1} yr^{-1}) could stimulate CH_4 uptake in the soil and that high fertilizer rates might inhibit the uptake. The methane monooxygenase can convert ammonia to nitrite, and high concentrations of ammonia will therefore reduce the ability of CH_4 uptake by methanotrophic bacteria. The mechanism is believed to be responsible for the inhibition of CH_4 uptake in the soil exposed to high concentrations of available nitrogen.

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However, in our study, increased fertilizer rates (0–400 kg N ha^{-1}) had no detectable effects on the cumulative CH_4 uptake by the soil in the wheat season. High fertilizer rates (>350 kg N ha^{-1}) tended to enhance the cumulative CH_4 uptake by the soil in the maize season. The increased CH_4 consumption by ammonia oxidizers, changes in the community composition of methanotrophic bacteria, general improvement of nutrient availability and enhanced CH_4 diffusivity through plants may have been responsible for the enhanced effects (Bodelier and Laanbroek, 2004). Furthermore, the enhanced effects of high fertilizer rates on CH_4 uptake by the soil were mainly observed within 1 to 4 days after the irrigation and continuous precipitation in the growing season for maize. An explanation for the delayed effect is that the upland soils may become partially anoxic after high precipitation and irrigation events and may start producing CH_4. We did observe weak CH_4 emissions (<25.1 µg C m^{-2} h^{-1}) after the continuous precipitation and irrigation events in the wheat-maize rotation field. The methanotrophic bacteria can benefit from the increased CH_4 concentration, and their population will increase when sufficient nitrogen is present (Bodelier and Laanbroek, 2004). After the soil dries, the methanotrophic bacteria may retain a high potential for atmospheric CH_4 consumption. Therefore, we observed delayed effects and enhanced effects of high fertilizer rates on atmospheric CH_4 uptake after the irrigation and continuous precipitation in the maize season in the wheat-maize rotation field.

4.2 Direct N_2O emission factors

Most of the global and national estimates of N_2O emissions from agricultural soils are based on the IPCC methodology of emission factors. However, the reported magnitude of direct N_2O emission factor ($EF_{F\text{-}N_2O}$) for chemical fertilizers is highly uncertain, and its variability may be attributed to the diversity of climate, soil, and field management and well as the period and frequency of flux measurement (Bouwman et al., 2002). Furthermore, fertilizer rates have increased rapidly in the last 15–20 yr, especially in developing countries, which has significantly influenced the magnitude of

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$EF_{F\text{-}N_2O}$. Therefore, more site-specific emission factors at an annual scale for different fertilizer rates are necessary for estimating global and national N_2O emissions from agricultural soils. In this study, we year-roundly measured the N_2O fluxes for the treatments of different fertilizer rates in a typical irrigated wheat-maize rotation field in northern China. The range of applied fertilizer rates (0–850 kg N ha^{-1} yr^{-1}) covered the spatial and temporal variability of fertilizer rate in the region. Because the relationship between the cumulative N_2O emissions and fertilizer rates could be described well by linear models, the calculated N_2O emission factors were constant for different nitrogen application rates (0.17 ± 0.03 % for the wheat season, 0.73 ± 0.07 % for the maize season and 0.49 ± 0.06 % for the annual scale). The estimated $EF_{F\text{-}N_2O}$ of 0.49 ± 0.06 % in the rotation year of 2009–2010 was slightly lower than the $EF_{F\text{-}N_2O}$ of 0.67 ± 0.23 % in the rotation year of 2008–2009 for the same experimental field (Liu et al., 2011). The direct N_2O emission factors for the wheat-maize rotation field in both rotation years were much lower than the global default $EF_{F\text{-}N_2O}$ of croplands (1.25 ± 1.0 %, IPCC, 1997; averaged 1.0 % for the croplands applied with mineral N-fertilizers, Bouwman et al., 2002; 1.0 %, IPCC, 2006). Ju et al. (2009) also reported a relatively low averaged $EF_{F\text{-}N_2O}$ of 0.12 and 0.23 % in the wheat and maize seasons in the wheat-maize rotation fields on the North China Plain. The results may suggest that the low available carbon sources and low WFPS for denitrification in most semi-humid upland soils in northern China are the reasons for the low $EF_{F\text{-}N_2O}$ (Ju et al., 2009, 2011).

4.3 Recommended rate of nitrogen fertilizer application

The fertilizer nitrogen rates of farmers' practice commonly range from 430 to 670 kg N ha^{-1} yr^{-1} for the typical wheat-maize rotation fields in northern China (Zhen et al., 2006; Ju et al., 2009; Liu et al., 2011). The recommended fertilizer rates by scientists are only in the range of 240 and 380 kg N ha^{-1} yr^{-1} (Liu et al., 2003; He et al., 2009; Ju et al., 2009; Wang et al., 2010). The overuse of nitrogen fertilizer of farmers' practice and the rapid increase of active nitrogen inputs by deposition and irrigation will not further increase the crop yield, but do cause a series of environmental problems (Ju et al., 2009). In this study, we can see the crop yield could not further increase if the annual fertilizer rate exceeded 650 kg N ha^{-1} (350 kg N ha^{-1} for maize and 300 kg N ha^{-1} for wheat). If the fertilizer rate increased from 150 to 250 and 350 kg N ha^{-1} for maize, the crop yield only increased 7 % and 20 % (0.5 and 1.5 t ha^{-1}) whereas the cumulative N_2O emissions increased 36 % and 111 % (0.72 and 2.21 kg N ha^{-1}). The high fertilizer rates for maize would especially increase the risk of N_2O emissions due to high temperatures, concentrated rainfall and irrigation between June and September. If the fertilizer rate increased from 120 to 180 and 300 kg N ha^{-1} for wheat, the crop yield only increased 1 % and 9 % (0.1 and 0.7 t ha^{-1}) whereas the cumulative N_2O emissions increased 34 % and 54 % (0.20 and 0.32 kg N ha^{-1}). Therefore, we recommend the local fertilizer rate should be controlled at approximately 120 kg N ha^{-1} for wheat and 150 kg N ha^{-1} for maize to reduce the risk of high N_2O emissions. The fertilizer rates for local farmers' use (430–470 kg N ha^{-1} yr^{-1}, Liu et al., 2011) can be reduced by 37–43 %. Then, the roughly estimated nitrogen inputs by fertilization (270 kg N ha^{-1} yr^{-1}), irrigation (about 4 kg N ha^{-1} yr^{-1}) and deposition (about 89 kg N ha^{-1} yr^{-1}, Ju et al., 2009; wet deposition: 30–43 kg N ha^{-1} yr^{-1}, this study and Liu et al., 2011) were approximately 363 ± 11 kg N ha^{-1} yr^{-1}. Applying the ratios of nitrogen losses reported by Ju et al. (2009), the processes of ammonia volatilization (approximately 60 kg N ha^{-1} yr^{-1} calculated as 19.4 and 24.7 % of applied nitrogen in the wheat and maize seasons), nitrate leaching (approximately 21 kg N ha^{-1} yr^{-1} calculated as 2.7 and 12.1 % of applied nitrogen in the wheat and maize seasons) and denitrification (approximately 5 kg N ha^{-1} yr^{-1} calculated as 0.1 and 3.3 % of applied nitrogen in the wheat and maize seasons) were estimated to lose 86 kg N ha^{-1} yr^{-1}. The harvested grain (wheat + maize) contained 262 ± 16 kg N ha^{-1} yr^{-1}. Accordingly, a slightly positive soil nitrogen balance (gain by 14 ± 27 kg N ha^{-1} yr^{-1}) could be achieved for the recommended fertilizer nitrogen rate to maintain the current crop yield, to keep the soil fertility and to reduce the N_2O emissions to the atmosphere.

Some studies have been conducted to investigate the effects of fertilizer nitrogen levels or optimized nutrient management on crop yield, nitrogen use efficiency, NO_3^-

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accumulation in deep soil and NO_3^- leaching in wheat-maize rotation fields in northern China (Liu et al., 2003; Zhao et al., 2006; He et al., 2009; Ju et al., 2009; Wang et al., 2010). The findings suggested that the fertilizer rates of farmers' practice could be reduced by 30 to 60 % to reduce the nitrogen losses to the environment and to maintain the current crop yield (5.8–7.7 t ha^{-1} for wheat and 7.1–8.9 t ha^{-1} for maize). The reported optimum fertilizer rates for the wheat-maize rotation fields in northern China varied between 120 and 180 kg N ha^{-1} for wheat and between 120 and 189 kg N ha^{-1} for maize. Our recommended fertilizer rates for wheat and maize were within these ranges.

5 Conclusions

We investigated the crop yield, N_2O and CH_4 fluxes for the treatments of six nitrogen levels in a full rotation year of 2009–2010 in a typical irrigated wheat-maize field in northern China. Our results suggest that the annual fertilizer rate should not exceed 650 kg N ha^{-1} yr^{-1} because higher fertilizer rates will not further increase the crop yield. The responses of cumulative N_2O emissions to fertilizer rates could be described well by linear models for the wheat season, maize season and annual scale, and the linear responses might be typical for the irrigated maize fields (Mosier et al., 2006; Halvorson et al., 2008). Therefore, the calculated N_2O emission factors (0.49 ± 0.06 % for the annual scale), which were considerably low as compared with the IPCC default 1 % (IPCC, 2006), were adaptive to the different fertilizer rates in the irrigated wheat-maize rotation field. Increased fertilizer rates had no detectable effects on the cumulative CH_4 uptake by the soil in the wheat season whereas very high fertilizer rates (≥350 kg N ha^{-1}) tended to enhance the CH_4 uptake in the maize season. The increase of crop yield induced by the increased fertilizer rates accompanied the exponential stimulation of N_2O emissions. A small increase in crop yield could be obtained when the fertilizer rate exceeded 120 kg N ha^{-1} for wheat and 150 kg N ha^{-1} for maize whereas the N_2O emissions steeply increased. We recommend the fertilizer rates

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for local farmers' use (430–470 kg N ha^{-1} yr^{-1}) to be reduced to 270 kg N ha^{-1} yr^{-1} to maintain the current high crop yield (>7 t ha^{-1} for maize and wheat) and to reduce the risk of superabundant nitrogen losses by ammonia volatilization, NO_3^- leaching and N_2O emissions (>3.51 kg N ha^{-1} yr^{-1}) in the wheat-maize rotation field.

Acknowledgements. This study was supported by the Ministry of Science and Technology of the P.R. China (2008BAD95B13) and the National Natural Science Foundation of China (41021004, 40711130636). Special thanks go to Guangren Liu, Yinghong Wang, Shixie Meng, Guangxuan Yan and Rui Wang for their technical support and help during field measurements.

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Table 1. Fertilizer rate, crop yield, nitrogen (N) content of grain, soil temperature (5 cm), water-filled pore space (WFPS, 0–6 cm), inorganic N and dissolved organic carbon (DOC) contents (0–10 cm) for the treatments of six fertilizer N levels.

Treatment	Fertilizer rate $kg\,N\,ha^{-1}$	Yield $t\,ha^{-1}$ $n=3^{*}$		Grain N content $g\,kg^{-1}$ $n=3^{*}$		Soil temperature °C $n=3^{**}$		WFPS % $n=5^{**}$		Inorganic N $mg\,N\,kg^{-1}$ SDW $n=4^{**}$		DOC $mg\,C\,kg^{-1}$ SDW $n=4^{**}$	
		WS	MS	WS	MS	WS	MS	WS	MS	WS	MS	WS	MS
N0	$0^{a}+0^{b}+0^{c}$	6.3^{A} (0.1)	4.9^{A} (0.7)	21.1^{A} (0.1)	12.0^{A} (0.5)	7.3^{A} (0.9)	23.1^{A} (0.6)	30.4^{A} (0.9)	48.9^{A} (1.6)	10.4^{A} (1.5)	5.2^{A} (0.7)	32.7^{A} (3.8)	47.4^{A} (5.0)
N135	$20^{a}+40^{b}+75^{c}$	6.6^{AB} (0.3)	6.9^{B} (0.3)	21.3^{A} (0.3)	12.7^{AB} (0.5)	8.4^{B} (0.8)	23.4^{BD} (0.6)	29.8^{B} (0.8)	49.1^{AB} (1.6)	22.5^{B} (2.7)	24.6^{B} (6.1)	33.7^{AB} (4.4)	66.1^{A} (10.1)
N270	$40^{a}+80^{b}+150^{c}$	7.5^{B} (0.4)	7.4^{BC} (0.6)	21.7^{AB} (0.3)	13.4^{ABC} (0.5)	7.6^{C} (0.9)	23.3^{B} (0.6)	30.5^{A} (0.8)	48.7^{A} (1.6)	26.7^{C} (3.6)	40.9^{C} (8.4)	36.4^{AB} (4.1)	70.7^{A} (10.1)
N430	$60^{a}+120^{b}+250^{c}$	7.6^{B} (0.4)	7.9^{BC} (0.6)	22.6^{B} (0.4)	13.7^{B} (0.2)	8.3^{B} (0.8)	23.0^{AC} (0.6)	30.5^{A} (0.8)	49.4^{BC} (1.7)	41.3^{D} (5.4)	72.3^{D} (18.0)	39.5^{B} (4.7)	81.7^{AB} (21.2)
N650	$100^{a}+200^{b}+350^{c}$	8.2^{B} (0.7)	8.9^{C} (0.7)	22.1^{AB} (0.4)	14.5^{BC} (0.8)	8.5^{B} (0.8)	22.9^{C} (0.6)	30.4^{A} (0.8)	49.1^{A} (1.7)	58.2^{E} (6.5)	91.6^{E} (21.9)	37.1^{B} (3.0)	94.5^{B} (32.5)
N850	$150^{a}+250^{b}+450^{c}$	7.5^{B} (0.5)	8.4^{BC} (0.8)	23.6^{AB} (1.6)	14.3^{C} (0.2)	7.4^{AC} (0.9)	23.5^{D} (0.5)	30.1^{AB} (0.8)	49.6^{C} (1.6)	63.8^{F} (6.8)	110.7^{F} (26.9)	34.1^{B} (3.3)	93.5^{B} (28.5)

Small letters a–c indicate the fertilization method and time: [a] tillage for 20 cm after surface broadcast at the wheat sowing time; [b] soil covering for 0–5 cm after band application at the turning-green stage of wheat; [c] soil covering for 0–5 cm after band application at the 18- to 19-leaf stage of maize. WS: the wheat growing season from 19 October 2009 to 15 June 2010. MS: the maize growing season from 16 June to 15 October 2010. *n*: spatial replicate number. Values in parentheses indicate standard errors. Different superscripts of capital letters indicate the significant differences between the treatments. * and ** indicate the significant differences at the levels of $p < 0.1$ and $p < 0.05$, respectively.

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Table 2. Minimum, maximum, averaged and cumulative CH_4 and N_2O fluxes for the treatments of six fertilizer nitrogen levels.

Gas	Treatment	Wheat season				Maize season				Annual	
		Min	Max	Ave	Cum	Min	Max	Ave	Cum	Ave	Cum.
		μg C or N m^{-2} h^{-1}			kg C or N ha^{-1}	μg C or N m^{-2} h^{-1}			kg C or N ha^{-1}	μg C of N m^{-2} h^{-1}	kg C or N ha^{-1}
CH_4	N0	−48.2	4.9	−16.9 (1.4)	1.01^{A} (0.05)	−98.5	3.6	−18.4 (3.3)	0.49^{A} (0.04)	−17.5 (1.6)	−1.49 (0.02)
	N135	−65.4	−4.6	−17.7 (1.5)	1.03^{A} (0.07)	−75.4	9.0	−21.1 (2.9)	0.57^{AB} (0.03)	−19.2 (1.5)	−1.59 (0.04)
	N270	−98.3	0.1	−18.9 (2.3)	1.16^{A} (0.03)	−96.3	7.5	−18.7 (3.5)	0.41^{AB} (0.06)	−18.8 (2.0)	−1.56 (0.04)
	N430	−56.4	25.1	−16.6 (1.6)	1.00^{A} (0.04)	−112.2	7.0	−22.8 (4.1)	0.52^{BC} (0.05)	−19.2 (2.0)	−1.51 (0.09)
	N650	−69.9	0.4	−15.7 (1.8)	0.97^{A} (0.09)	−132.5	−9.0	−28.6 (4.4)	0.71^{D} (0.00)	−21.2 (2.2)	−1.69 (0.10)
	N850	−74.3	5.6	−16.7 (2.0)	1.09^{A} (0.01)	−158.4	−3.1	−25.4 (4.3)	0.70^{C} (0.06)	−20.5 (2.2)	−1.79 (0.07)
N_2O	N0	1.1	59.1	10.4 (1.5)	0.48^{A} (0.11)	9.8	68.4	32.6 (2.4)	1.01^{A} (0.15)	19.9 (1.7)	1.50 (0.26)
	N135	2.1	26.8	9.2 (0.8)	0.48^{A} (0.07)	6.8	424.2	82.1 (17.1)	1.61^{B} (0.14)	40.3 (8.1)	2.09 (0.20)
	N270	1.9	44.5	13.4 (1.4)	0.59^{AB} (0.11)	9.2	640.6	104.5 (23.1)	2.00^{B} (0.27)	52.3 (10.9)	2.59 (0.33)
	N430	1.8	66.2	17.7 (1.8)	0.79^{B} (0.10)	4.8	718.3	153.7 (33.2)	2.72^{C} (0.16)	75.8 (15.7)	3.51 (0.26)
	N650	1.7	65.2	20.0 (2.3)	0.91^{B} (0.25)	12.4	954.3	228.4 (45.4)	4.21^{D} (0.57)	109.0 (22.0)	5.12 (0.78)
	N850	3.1	110.0	25.6 (3.2)	1.08^{B} (0.11)	13.8	1017.8	235.3 (46.2)	4.49^{D} (0.58)	115.1 (22.4)	5.57 (0.68)

Different superscripts of capital letters indicate the significant differences at the level of $p < 0.05$ between the treatments. Values in parentheses indicate standard error of the seasonal and annual averages.

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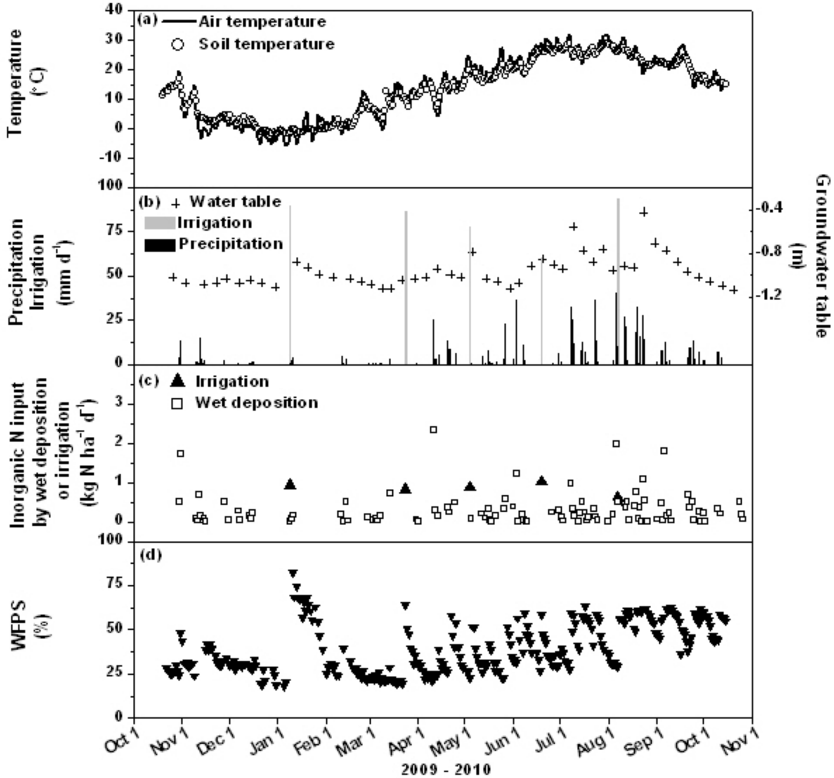

Fig. 1. Temporal courses of **(a)** air and soil temperatures, **(b)** daily precipitation, irrigation and groundwater table, **(c)** inorganic nitrogen (N) inputs by irrigation and wet deposition and **(d)** soil water-filled pore space (WFPS) at the experimental field.

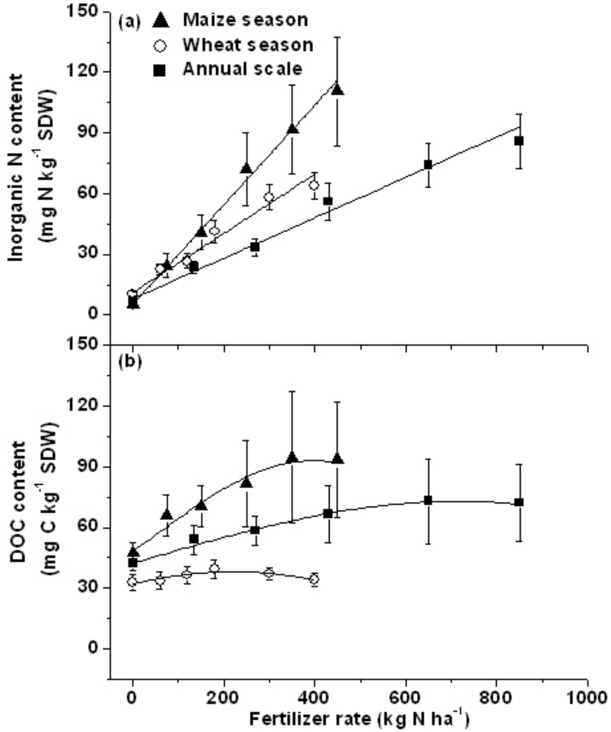

Fig. 2. Average contents of soil inorganic nitrogen (N) and dissolved organic carbon (DOC) against the fertilizer N rates. SDW: soil dry weight. Error bars represent standard error of the seasonal or annual averages. Fitting equations for annual scale: **(a)** $y = 8.08 + 0.10 \cdot x$ ($r^2 = 0.99$, $p < 0.01$); **(b)** $y = \exp(3.75 + 0.002 \cdot x - 0.000001 \cdot x^2)$ ($r^2 = 0.97$, $p < 0.01$); for wheat season: **(a)** $y = 11.09 + 0.15 \cdot x$ ($r^2 = 0.97$, $p < 0.01$); **(b)** $y = \exp(3.47 + 0.002 \cdot x - 0.000004 \cdot x^2)$ ($r^2 = 0.81$, $p < 0.05$); for maize season: **(a)** $y = 5.22 + 0.25 \cdot x$ ($r^2 = 0.99$, $p < 0.01$); **(b)** $y = \exp(3.88 + 0.003 \cdot x - 0.000004 \cdot x^2)$ ($r^2 = 0.94$, $p < 0.01$).

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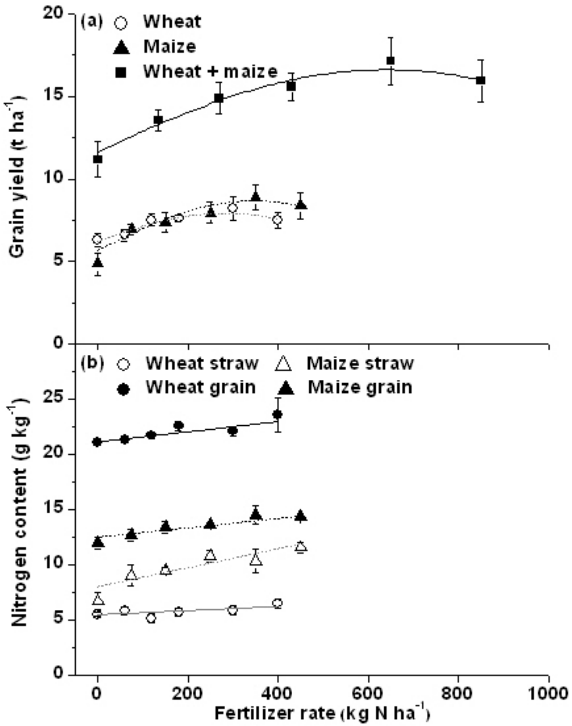

Fig. 3. Crop yield, nitrogen contents in crop straw and grain against the fertilizer rates. Error bars represent standard error of the treatment averages. Fitting equations: **(a)** for wheat: $y = \exp(1.82 + 0.002 \cdot x - 0.000003 \cdot x^2)$ ($r^2 = 0.89$, $p < 0.01$); for maize: $y = \exp(1.73 + 0.002 \cdot x - 0.000004 \cdot x^2)$ ($r^2 = 0.78$, $p < 0.05$); for wheat and maize: $y = \exp(2.45 + 0.001 \cdot x - 0.000009 \cdot x^2)$ ($r^2 = 0.93$, $p < 0.01$); **(b)** for wheat straw: $y = 5.52 + 0.002 \cdot x$ ($r^2 = 0.73$, $p < 0.05$); for wheat grain: $y = 21.15 + 0.005 \cdot x$ ($r^2 = 0.74$, $p < 0.05$); for maize straw: $y = 8.03 + 0.009 \cdot x$ ($r^2 = 0.80$, $p < 0.05$); for maize grain: $y = 12.51 + 0.004 \cdot x$ ($r^2 = 0.90$, $p < 0.01$).

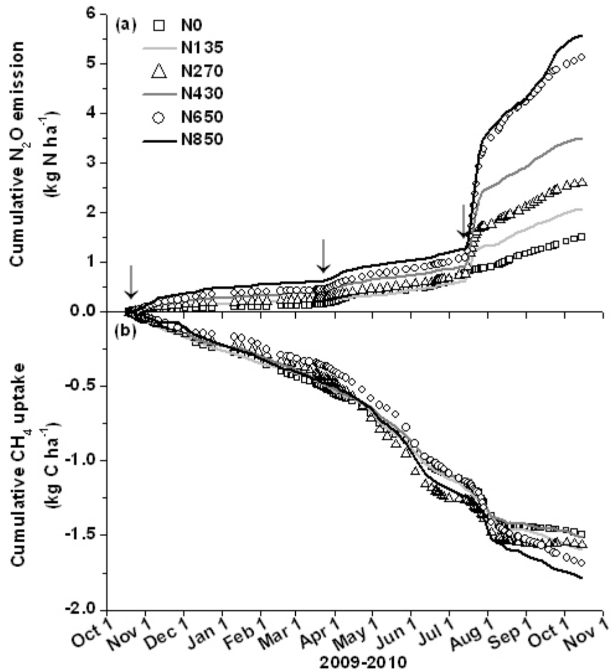

Fig. 4. Temporal courses of cumulative **(a)** N_2O emissions and **(b)** CH_4 uptake for the treatments of six nitrogen levels (0, 135, 270, 430, 650 and 850 kg N ha^{-1} yr^{-1}). Each data point is the arithmetic mean of fluxes measured at three spatial replicates. The arrows indicate the fertilization dates.

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Responses of N_2O and CH_4 fluxes to fertilizer nitrogen addition rates

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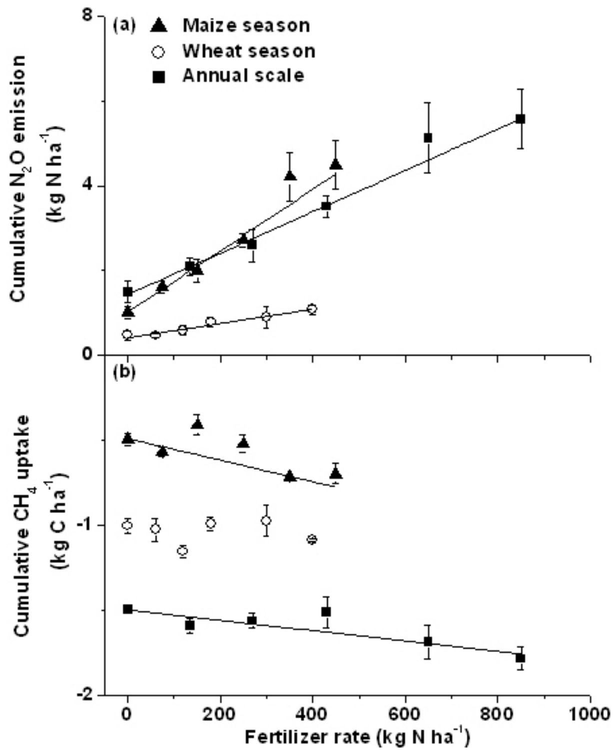

Fig. 5. Relationship between cumulative N_2O emissions, cumulative CH_4 uptake and addition rates of nitrogen fertilizer. Error bars represent standard error of the treatment averages. Fitting equations for annual scale: **(a)** $y = 1.44 + 0.005 \cdot x$ ($r^2 = 0.99$, $p < 0.01$); **(b)** $y = -1.50 - 0.0003 \cdot x$ ($r^2 = 0.82$, $p < 0.05$); for wheat season: **(a)** $y = 0.41 + 0.002 \cdot x$ ($r^2 = 0.94$, $p < 0.01$); for maize season: **(a)** $y = 1.01 + 0.007 \cdot x$ ($r^2 = 0.97$, $p < 0.01$); **(b)** $y = -0.49 - 0.0006 \cdot x$ ($r^2 = 0.77$, $p < 0.05$).

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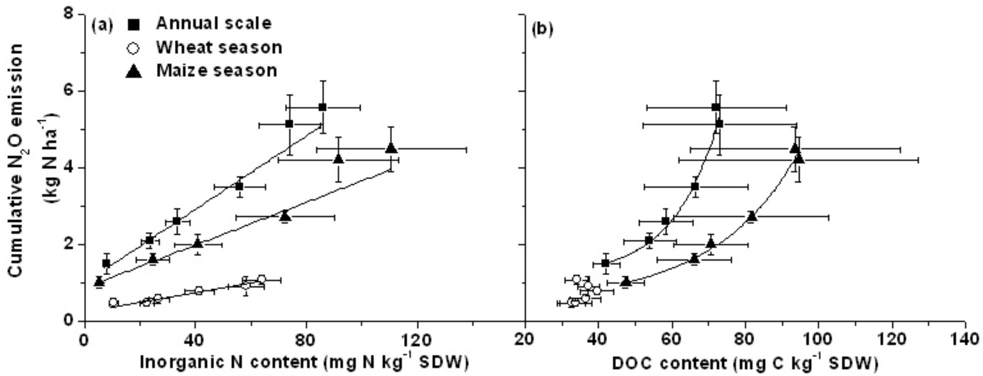

Fig. 6. Relationship between cumulative N_2O emissions and average soil contents of inorganic nitrogen (N) and dissolved organic carbon (DOC). SDW: soil dry weight. Error bars for vertical and horizontal ordinates represent standard error of the treatment averages and annual averages, respectively. Fitting equations for annual scale: **(a)** $y = 1.00 + 0.05 \cdot x$ ($r^2 = 0.97$, $p < 0.01$); **(b)** $y = \exp(1.11 - 0.05 \cdot x + 0.0008 \cdot x^2)$ ($r^2 = 0.97$, $p < 0.01$); for wheat season: **(a)** $y = 0.25 + 0.01 \cdot x$ ($r^2 = 0.92$, $p < 0.01$); for maize season: **(a)** $y = 0.87 + 0.03 \cdot x$ ($r^2 = 0.95$, $p < 0.01$); **(b)** $y = \exp(-0.69 + 0.006 \cdot x + 0.0002 \cdot x^2)$ ($r^2 = 0.99$, $p < 0.01$).

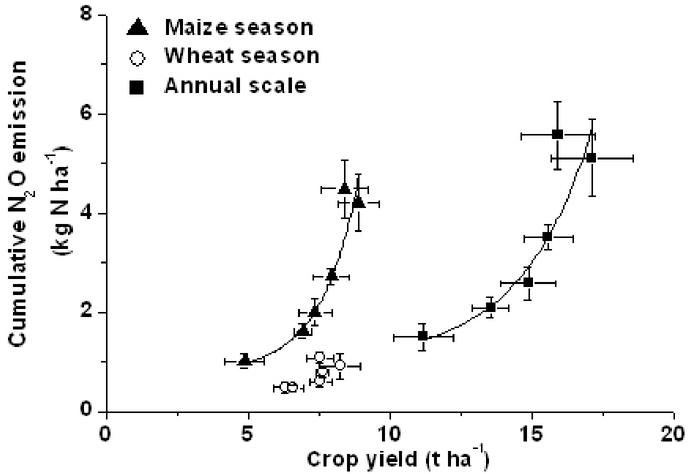

Fig. 7. Relationship between cumulative N_2O emissions and crop yield. Error bars represent standard error of the treatment averages. Fitting equation for annual scale: $y = \exp(1.41 - 0.31 \cdot x + 0.02 \cdot x^2)$ ($r^2 = 0.80$, $p < 0.05$); for maize season: $y = \exp(0.96 - 0.52 \cdot x + 0.07 \cdot x^2)$ ($r^2 = 0.93$, $p < 0.01$).

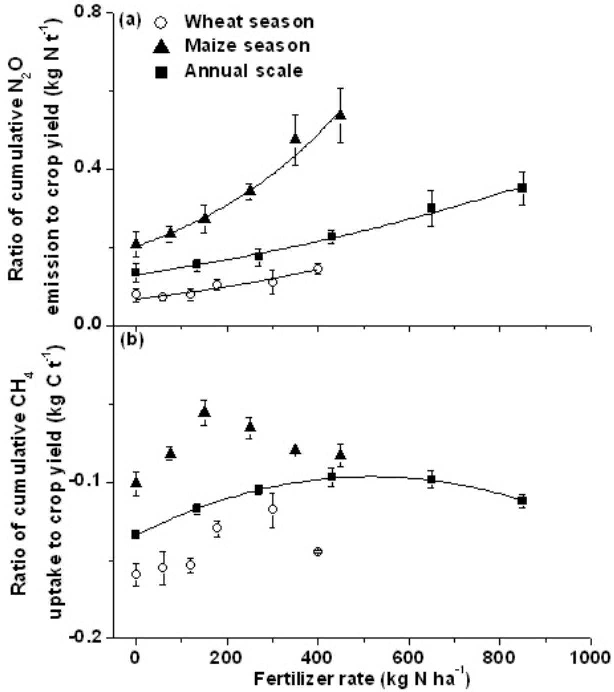

Fig. 8. Ratio of cumulative N_2O emissions and CH_4 uptake to crop yield across the nitrogen addition gradient. Error bars represent standard error of the treatment averages. Fitting curves for annual scale: **(a)** $y = \exp(-2.05 + 0.001 \cdot x - 0.0000002 \cdot x^2)$ ($r^2 = 0.99$, $p < 0.01$); **(b)** $y = -0.13 + 0.0001 \cdot x - 0.0000001 \cdot x^2$ ($r^2 = 0.999$, $p < 0.01$); for wheat season: **(a)** $y = \exp(-2.70 + 0.002 \cdot x + 0.0000002 \cdot x^2)$ ($r^2 = 0.91$, $p < 0.01$); for maize season: **(a)** $y = \exp(-1.60 + 0.002 \cdot x + 0.0000005 \cdot x^2)$ ($r^2 = 0.98$, $p < 0.01$).

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