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Responses of N₂O and CH₄ fluxes to fertilizer nitrogen addition rates in an irrigated wheat-maize cropping system in northern China

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Abstract. Model and field studies generally posit that when the application rates of nitrogen fertilizer exceed crop needs, nitrous oxide (N2O) emissions will increase nonlinearly, though linear responses are also extensively reported by field studies. We conducted year-round measurements of crop yield, N2O and methane (CH4) fluxes for treatments of six fertilizer nitrogen levels (0, 135, 270, 430, 650 and $850 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in the form of urea) in a typical irrigated wheat-maize rotation field in northern China. Linear models characterized the responses of cumulative N₂O emissions to fertilizer rates well; therefore, the calculated N_2O emission factors of $0.17 \pm 0.02\%$, $0.73 \pm 0.05\%$ and 0.49 ± 0.02 % for the wheat season, maize season and annual scale, respectively, were appropriate for the different fertilizer rates. The cumulative CH₄ uptake by the soil tended to be enhanced at higher fertilizer rates ($>350 \text{ kg N ha}^{-1}$) in the maize season whereas no effect was observed for the wheat season. When the annual fertilizer rates increased from 270 to 430, from 270 to 650, and from 270 to $850 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$, the crop yields increased only $3 \sim 15\%$ (0.5 $\sim 2.1 \, \text{t ha}^{-1} \, \text{yr}^{-1}$), but cumulative N₂O emissions increased $35 \sim 115 \% (0.9 \sim 3.0 \text{ kg N ha}^{-1} \text{ yr}^{-1})$. We recommend 270 kg N ha⁻¹ yr⁻¹as the locally optimum fertilizer rate. Considering the nitrogen inputs by fertilization $(270 \text{ kg N ha}^{-1} \text{ yr}^{-1})$, irrigation $(4.3 \pm 0.2 \text{ kg N ha}^{-1} \text{ yr}^{-1})$ and deposition (wet deposition: $30.5 \pm 1.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), the slightly positive soil nitrogen balance could maintain the current crop yield (\sim 13.8 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 $> 430 \text{ kg N ha}^{-1} \text{ yr}^{-1}$).

1 Introduction

Methane (CH₄) and nitrous oxide (N₂O) are considered the most potent greenhouse gases after carbon dioxide (CO₂). Their global warming potential indices relative to the reference gas CO₂ are 25 and 298, respectively, on a 100-year time horizon. N₂O also participates in the destruction of stratospheric ozone (O₃) (Forster et al., 2007). Fertilized upland agricultural soils are the most important anthropogenic N₂O sources and are significant CH₄ sinks (Bouwman et al., 2002; Dutaur and Verchot, 2007). The production of N₂O and the consumption of CH4 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 CH₄ and N₂O 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 $(>100 \text{ kg N ha}^{-1} \text{ yr}^{-1})$, may inhibit CH₄ 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 CH₄ 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 N2O emissions from cultivated soils. The relationship between N2O emissions and fertilizer rates is complex. The response curves of N₂O 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 N₂O 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, N₂O emissions are stimulated (McSwiney and Robertson, 2005; Zebarth et al., 2008). Some studies reported the N₂O 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 for different years and sites with on-farm maize crops in the US Midwest. Using a 5yr field measurement, Mosier et al. (2006) and Halvorson et al. (2008) reported that the N₂O 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, http://www.stats. gov.cn/english/statisticaldata/). 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 \sim 690 mm yr $^{-1}$, Wang et al., 2008) and synthetic fertilizers (550 \sim 600 kg N ha⁻¹ yr⁻¹, Ju et al., 2009). With a rapid increase in the fertilizer rate, the use efficiencies of synthetic fertilizers evaluated as yield per unit synthetic fertilizer sharply decrease (Tong et al., 2003; Ju et al., 2009). The overuse of synthetic nitrogen fertilizers creates many environmental problems, such as nitrate leaching, water pollution, soil salination, soil acidification and additional N₂O emissions (Zhen et al., 2006; Ju et al., 2009). Until now, the responses of N₂O emissions, CH₄ uptake and crop yield to increased fertilizer rates in the wheat-maize rotation fields have remained unclear. Studying the responses of N₂O emissions, CH₄ uptake and crop yield to fertilizer rates may help to explain the applicability of a set emission factors for N2O to different fertilizer rates and may help to determine the optimum fertilizer rate for reducing N₂O emissions, enhancing the CH₄ 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, N2O and CH4 fluxes to fertilizer rates; (b) quantify the emission factors of N₂O for different fertilizer rates; and (c) determine the relationships between N₂O/CH₄ fluxes and crop yield and recommend the optimum fertilizer rate for the wheat-maize rotation fields.

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 Mottlic Hapli-Ustic Argosols (Gong et al., 2007), which was analogue to the Luvisols (IUSS Working Group WRB, 2006; Krasilnikov et al., 2009). Soil properties for the uppermost 10 cm were clay content $31.8 \pm 0.9\%$, silt content $38.9 \pm 0.6\%$, sand content 29.3 ± 1.4 %, organic carbon content 11.3 ± 0.6 g kg⁻¹, total nitrogen content 1.12 ± 0.05 g kg⁻¹ and pH 8.7. The soil has a bulk density of $1.17 \pm 0.04 \,\mathrm{g \, cm^{-3}}$ (mean value \pm standard error, $0 \sim 6$ cm). The history of the management of the experimental field is provided in Liu et al. (2011).

Eighteen experimental plots $(6 \times 6 \text{ m each})$ with three replicates of six fertilizer nitrogen levels (0, 135, 270, 430, 650 and $850 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$, 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 $20 \sim 23$ -cm row spacing on 19 October 2009, and with summer maize (Zea mays L.) at 60-cm row spacing and $20 \sim 22$ -cm plant spacing 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 $5 \sim 10 \, \mathrm{cm}$ after harvest and was mechanically ploughed into the soil $(0 \sim 20 \,\mathrm{cm})$ just before seeding. A motor-pumped well beside the experimental field was used to pump underground water (depth: $130 \sim 140 \,\mathrm{m}$) to a manually movable sprinkler irrigation system and irrigate the crops. Herbicide (atrazine) was applied once each wheat and maize growing season. Pesticides (the mixture of emamectin benzoate and chlorpyrifos) were 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 $0 \sim 5$ cm after band application) and at the 18- to 19-leaf stage of maize (soil covering for $0 \sim 5$ cm after band application) (Table 1). In addition, the calcium superphosphate $(60.0 \text{ kg P ha}^{-1} \text{ yr}^{-1})$ and potassium sulfate $(30.0 \text{ kg K ha}^{-1} \text{ yr}^{-1})$ were applied at the wheat sowing time for all treatments.

2.2 Measurement of N₂O and CH₄ 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

Table 1. Fertilizer rate, crop yield, aboveground biomass (AB), nitrogen contents of grain (GN) and aboveground plant (APN), soil temperature (ST, 5 cm), soil water-filled pore space (WFPS, $0 \sim 6$ cm), soil inorganic nitrogen (IN, $0 \sim 10$ cm) and dissolved organic carbon contents (DOC, $0 \sim 10$ cm) for the treatments of six fertilizer nitrogen levels.

	Fertilizer rate	Crop yield $t ha^{-1}$ $n = 3^*$		AB t ha ⁻¹		GN kg N ha ⁻¹		APN kg N ha ⁻¹		°C		WFPS %		IN mg N kg ⁻¹ SDW		mg C kg ⁻¹ SDW	
Treatment	kg N ha−1																
				n =	n = 3*		n = 3*		n = 3*		n = 3**		n = 5**		n = 4**		n = 4**
		WS	MS	WS	MS	WS	MS	WS	MS	WS	MS	WS	MS	WS	MS	WS	MS
N0	$0^a + 0^b + 0^c$	6.3 ^A	4.9 ^A	15.0 ^A	13.7 ^A	133.1 ^A	58.4 ^A	180.8 ^A	117.3 ^A	7.3 ^A	23.1 ^A	30.4 ^A	48.9 ^A	10.4 ^A	5.2 ^A	32.7 ^A	47.4 ^A
		(0.1)	(0.7)	(0.1)	(0.7)	(2.7)	(8.5)	(3.3)	(10.5)	(0.9)	(0.6)	(0.9)	(1.6)	(1.5)	(0.7)	(3.8)	(5.0)
N135	$20^a + 40^b + 75^c$	6.6^{AB}	5.9 ^{AB}	15.4 ^{AB}	14.7 ^{AB}	140.9 ^{AB}	74.9 ^{AB}	191.7 ^{AB}	151.3 ^{AB}	8.4^{B}	23.4^{BD}	29.8^{B}	49.1 ^{AB}	22.5^{B}	24.6^{B}	33.7 ^{AB}	66.1 ^A
		(0.3)	(0.9)	(0.6)	(1.8)	(7.6)	(11.7)	(8.4)	(17.1)	(0.8)	(0.6)	(0.8)	(1.6)	(2.7)	(6.1)	(4.4)	(10.1)
N270	$40^a + 80^b + 150^c$	7. ^B	6.8^{AB}	15.5 ^{AB}	16.9 ^{AC}	151.3 ^B	91.6 ^{AB}	200.8^{AB}	180.9 ^{AC}	7.6 ^C	23.3^{B}	30.5^{A}	48.7 ^A	26.7 ^C	40.9 ^C	36.4 ^{AB}	70.7 ^A
		(0.3)	(0.8)	(0.3)	(1.6)	(6.9)	(11.0)	(7.6)	(12.9)	(0.9)	(0.6)	(0.8)	(1.6)	(3.6)	(8.4)	(4.1)	(10.1)
N430	$60^a + 120^b + 250^c$	7.3^{B}	7.0^{AB}	16.8 ^B	18.0 ^{BC}	164.3 ^B	95.4 ^{AB}	218.7^{B}	208.7 ^{BC}	8.3^{B}	23.0 ^{AC}	30.5^{A}	49.4 ^{BC}	41.3 ^D	72.3 ^D	39.5 ^B	81.7 ^{AB}
		(0.4)	(1.0)	(0.9)	(1.6)	(9.9)	(15.9)	(10.9)	(17.4)	(0.8)	(0.6)	(0.8)	(1.7)	(5.4)	(18.0)	(4.7)	(21.2)
N650	$100^a + 200^b + 350^c$	8.2^{B}	7.6 ^{AB}	17.7 ^B	19.1 ^C	181.9 ^B	110.0^{AB}	237.7^{B}	223.2 ^C	8.5^{B}	22.9 ^C	30.4 ^A	49.1 ^A	58.2 ^E	91.6 ^E	37.1^{B}	94.5 ^B
		(0.7)	(1.1)	(1.3)	(1.2)	(16.3)	(22.9)	(17.2)	(26.4)	(0.8)	(0.6)	(0.8)	(1.7)	(6.5)	(21.9)	(3.0)	(32.5)
N850	$150^a + 250^b + 450^c$	7.5^{B}	8.4 ^B	16.5 ^{AB}	21.1 ^C	177.8 ^B	120.3^{B}	236.2 ^{AB}	260.2 ^C	7.4 ^{AC}	23.5^{D}	30.1^{AB}	49.6 ^C	63.8 ^F	110.7 ^F	34.1^{B}	93.5 ^B
		(0.5)	(0.8)	(1.1)	(1.5)	(16.2)	(11.7)	(17.1)	(14.5)	(0.9)	(0.5)	(0.8)	(1.6)	(6.8)	(26.9)	(3.3)	(28.5)

Small letters a \sim c indicate the fertilization method and time: ^a tillage for 20 cm after surface broadcast at the wheat sowing time; ^b soil covering for $0\sim$ 5 cm after band application at the turning-green stage of wheat; ^c soil covering for $0\sim$ 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 June 16 to October 15, 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.

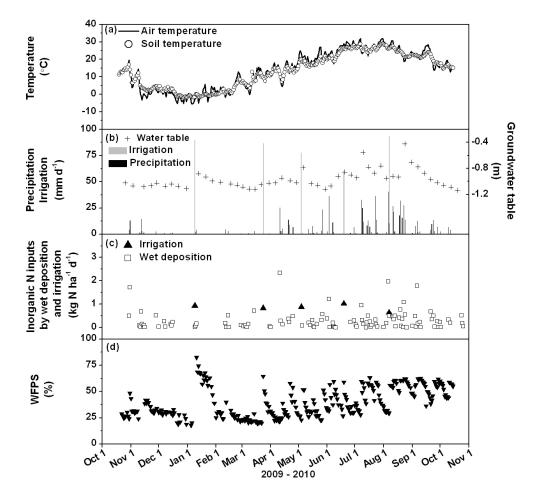


Fig. 1. Temporal courses of (a) air and soil temperatures, (b) daily precipitation, irrigation and groundwater table, (c) inorganic nitrogen (N) inputs by irrigation and wet deposition, and (d) soil water-filled pore space (WFPS) at the experimental field.

static chamber-gas chromatograph measuring system as described in Yao et al. (2009). Prior to seeding, two types of base collar (length \times width = 50×50 and 50×30 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 \times width \times height = $50 \times 50 \times 50$ 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 \times width \times height = $50 \times 30 \times 20$ cm), separated it vertically into two parts and drilled a hole (diameter: 11 cm) in the center of chamber top. The local planting density of maize varied between 6.5 and 7.5 plants m⁻² (equal to $0.13 \sim 0.16 \,\mathrm{m}^2$ per plant). The cross sectional area of type II chamber was $0.15 \,\mathrm{m}^2$, and therefore it could cover the soil surface area representatively. 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 preservative film (thickness: 1.2 µm, material: polyvinylidene chloride, oxygen permeability: $2.9 \,\mathrm{cm}^3 \,\mathrm{m}^{-2} \,\mathrm{h}^{-1} \,\mathrm{atm}^{-1}$) when the chamber was closed. The preservative film was wrapped 3 layers to reduce the leaking of gases through it. 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 $20 \sim 40 \,\mathrm{min}$. Five gas samples in total were taken and stored in the plastic syringes. The CH₄ and N₂O concentrations of gas samples were analyzed within 10 hours after sampling by a gas chromatograph (GC, Agilent 5890, Agilent Technologies Inc., USA), which was equipped with an electron capture detector (ECD) and a flame ionization detector (FID). The ECD, FID and column of the GC were heated to 330, 200 and 55 °C, respectively. The carrier gas of GC was N2 (99.999 % purity). To avoid interference of CO₂ of the gas samples with the ECD-cell during N2O detection, the make-up gas contained a high concentration of CO₂ (approximately 10 % CO₂ in N₂, Zheng et al. 2008). The CH₄ and N₂O concentrations were hourly calibrated by three reference gas injections (2.10 ppm CH₄ and 1.02 ppm N₂O in N₂, Air Products and Chemicals, Inc., Beijing, China). The flux calculation was based on the nonlinear fitting approach, as described by Liu et al. (2010). Sampling at $08:00 \sim 10:00$ a.m. local time, resulted in the lowest deviation $(5.3 \sim 8.6 \%)$ 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.

2.3 Auxiliary measurements

In addition to flux measurements, we also measured crop yield, carbon and nitrogen contents of crop straw and grain, air temperature, air pressure, precipitation, groundwater table, soil temperature, soil moisture, soil ammonium (NH_4^+) , nitrate (NO₃⁻) and dissolved organic carbon (DOC) contents. At harvest, three replicates (0.36 m² each for wheat and 2 m² 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 $(0 \sim 6 \, \text{cm})$ 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 DOC and inorganic nitrogen $(NH_4^+ + NO_3^-)$ contents of the topsoil $(0 \sim 10 \, \text{cm})$ were measured from four randomly collected soil samples for each nitrogen level, either once per $1 \sim 2$ days for two 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, NH₄⁺ and NO₃⁻ analyzer (San⁺⁺ Continuous Flow Analyzer, Skalar Analytical B.V., The 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^+

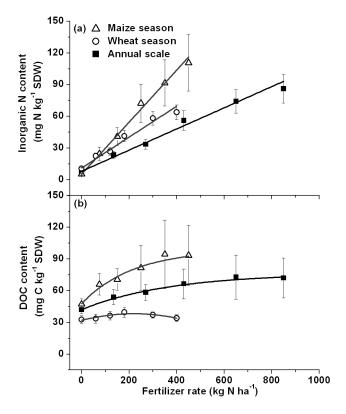


Fig. 2. Average contents of soil inorganic nitrogen (N) and dissolved organic carbon (DOC) against the fertilizer rates. SDW: soil dry weight. Error bars represent standard error of the seasonal or annual averages.

and NO_3^- contents between treatments. Nonparametric test of two independent samples (Mann-Whitney U-test) was used to analyze the significance of the differences in the mean crop yield, aboveground biomass, nitrogen contents of grain and aboveground plant between treatments. A t-test and an F-test were performed to determine the significance of regression coefficients and regression equations, respectively. When the probability value of t-test and F-test for the significance of regression coefficients and equations <0.05, the regression equations were selected 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.

3 Results

3.1 Environment, soil inorganic nitrogen and DOC

The annual mean air and soil (5 cm) temperatures at the experimental field were 13.8 ± 0.6 and 13.4 ± 0.5 °C (\pm s.e.), respectively (Fig. 1a). The annual total precipitation was 666.2 mm, which was 19 % higher than the annual mean during the most recent 10 yr. The total amount of irrigation

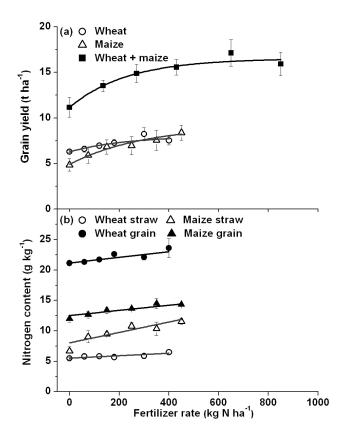


Fig. 3. Crop yield, nitrogen contents in crop straw and grain against the fertilizer rates. Error bars represent standard error of the treatment averages.

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 $(0\sim6$ 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 nitrogen fertilizer 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,

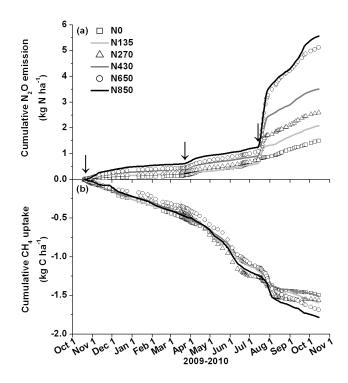


Fig. 4. Temporal courses of cumulative (**a**) N_2O emissions and (**b**) CH₄ uptake for the treatments of six nitrogen levels (0, 135, 270, 430, 650 and 850 kg N ha⁻¹ yr⁻¹). Each data point is the arithmetic mean of fluxes measured at three spatial replicates. The arrows indicate the fertilization dates.

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 and Table 3).

The soil (0 \sim 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 \sim 24 mg C kg⁻¹ SDW) whereas the maximum values (161 \sim 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 and Table 3).

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 tha⁻¹ for wheat and

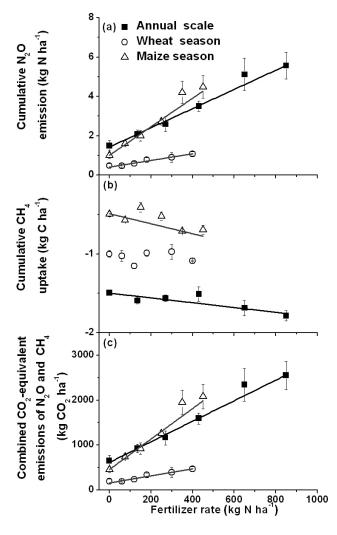


Fig. 5. Relationship between cumulative N_2O emissions, cumulative CH_4 uptake, combined CO_2 -equivalent emissions of N_2O and CH_4 and fertilizer rates (calculated as 298 and 25 CO_2 -equivelents for N_2O and CH_4 on a 100-yr time horizon). Error bars represent standard error of the treatment averages.

from 4.9 ± 0.7 to 8.4 ± 0.8 t ha⁻¹ for maize (Table 1). The maximum crop yields were obtained at the fertilizer rate of $300 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1}$ for wheat and $450 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1}$ for maize (Table 1 and Fig. 3a). The relationships of crop yield and fertilizer rate could be characterized by exponentially fitting curves (p < 0.01, Fig. 3a and Table 3). The increased fertilizer rate also significantly increased the nitrogen contents of harvested crop straw and grain, especially for the maize straw (p < 0.05, Fig. 3b and Table 3).

3.3 N₂O and CH₄ fluxes

The N_2O fluxes for N_0 varied between 1.1 and $68.4 \,\mu g \, N \, m^{-2} \, h^{-1}$, with an annual mean of $19.9 \pm 1.7 \,\mu g \, N \, m^{-2} \, h^{-1}$. The relatively high fluxes (>40 $\,\mu g \, N \, m^{-2} \, h^{-1}$) for N_0 were observed after the

Table 2. Minimum, maximum, averaged and cumulative CH₄ and N₂O fluxes for the treatments of six fertilizer nitrogen levels.

				Wheat season				Maize season	Annual			
Gas	Treatment	Min	Max	Ave	Cum	Min	Max	Ave	Cum	Ave	Cum	
		μg C or N m ⁻² h ⁻¹			kg C or N ha ⁻¹	μд	C or N m	$^{-2} h^{-1}$	kg C or N ha ⁻¹	$\mu g \ C \ of \ N \ m^{-2} \ h^{-1}$	${\rm kg}~{\rm C}~{\rm or}~{\rm N}~{\rm ha}^{-1}$	
	N0	-48.2	4.9	-16.9 (1.4)	$1.00^{A} (0.05)$	-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)	1.02 ^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)	1.15^{A} (0.03)	-96.3	7.5	-18.7(3.5)	0.41^{AB} (0.06)	-18.8(2.0)	-1.56(0.04)	
CH_4	N430	-56.4	25.1	-16.6(1.6)	0.99 ^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 (0.004)	-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)	
	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)	
N_2O	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.

Table 3. Fitting equations for Figs. 2, 3, 5, 6, 7 and 8.

Figure		Fitting equation	r^2	p	Figure		Fitting equation	r^2	p
Fig. 2a	WS:	$f(x) = 11.09 + 0.15 \cdot x$	0.97	< 0.01	Fig. 2b	WS:	$f(x) = \exp(3.47 + 0.002 \cdot x - 0.000004 \cdot x^2)$	0.81	< 0.05
	MS:	$f(x) = 5.22 + 0.25 \cdot x$	0.997	< 0.01		MS:	$f(x) = 98.85 - 51.19 \cdot 0.995^x$	0.97	< 0.01
	AS:	$f(x) = 8.08 + 0.10 \cdot x$	0.99	< 0.01		AS:	$f(x) = 76.41 - 34.21 \cdot 0.997^x$	0.98	< 0.01
Fig. 3a	Wheat:	$f(x) = 8.03 - 1.74 \cdot 0.996^x$	0.90	< 0.01	Fig. 3b	Wheat straw:	$f(x) = 5.49 + 0.002 \cdot x$	0.79	< 0.05
-	Maize:	$f(x) = 9.16 - 4.24 \cdot 0.997^x$	0.96	< 0.01	_	Wheat grain:	$f(x) = 21.15 + 0.005 \cdot x$	0.74	< 0.05
	Wheat + Maize:	$f(x) = 16.51 - 5.36 \cdot 0.996^x$	0.95	< 0.01		Maize straw:	$f(x) = 8.03 + 0.009 \cdot x$	0.80	< 0.05
						Maize grain:	$f(x) = 12.51 + 0.004 \cdot x$	0.90	< 0.01
Fig. 5a	WS:	$f(x) = 0.41 + 0.0017 \cdot x$	0.94	< 0.01	Fig. 5b	MS:	$f(x) = -0.49 - 0.0006 \cdot x$	0.77	< 0.05
-	MS:	$f(x) = 1.01 + 0.0073 \cdot x$	0.97	< 0.01		AS:	$f(x) = -1.50 - 0.0003 \cdot x$	0.82	< 0.05
	AS:	$f(x) = 1.44 + 0.0049 \cdot x$	0.99	< 0.01					
Fig. 5c	WS:	$f(x) = 159.93 + 0.79 \cdot x$	0.94	< 0.01	Fig. 6a	WS:	$f(x) = 0.25 + 0.01 \cdot x$	0.92	< 0.01
	MS:	$f(x) = 457.80 + 3.39 \cdot x$	0.97	< 0.01		MS:	$f(x) = 0.87 + 0.03 \cdot x$	0.95	< 0.01
	AS:	$f(x) = 619.19 + 2.28 \cdot x$	0.99	< 0.01		AS:	$f(x) = 1.00 + 0.05 \cdot x$	0.97	< 0.01
Fig. 6b	MS:	$f(x) = 0.20 \cdot 1.03^x$	0.98	< 0.01	Fig.7	WS:	$f(x) = \exp(-4.29 + 0.55 \cdot x)$	0.69	< 0.05
-	AS:	$f(x) = 0.21 \cdot 1.04^x$	0.94	< 0.01	_	MS:	$f(x) = 0.12 \cdot 1.56^x$	0.93	< 0.01
						AS:	$f(x) = \exp(-2.50 + 0.24 \cdot x)$	0.83	< 0.05
Fig. 8a	WS:	$f(x) = 0.07 \cdot 1.002^x$	0.90	< 0.01	Fig. 8b	AS	$f(x) = -0.14 + 0.0001^* \cdot x - 0.000000009 \cdot x^2$	0.95	< 0.01
-	MS:	$f(x) = 0.22 \cdot 1.002^x$	0.93	< 0.01	_				
	AS:	$f(x) = 0.14 \cdot 1.001^x$	0.96	< 0.01					

r: correlation coefficient; p: probability value; WS: wheat season; MS: maize season; AS: annual scale.

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 $\mu g\,N\,m^{-2}\,h^{-1}$ for N135, N270, N430, N650 and N850, respectively. The extremely high emissions (>200 $\mu g\,N\,m^{-2}\,h^{-1}$) 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 $115.1\pm22.4\,\mu g\,N\,m^{-2}\,h^{-1}$ (Table 2).

The CH_4 fluxes fluctuated between -158.4 and $25.1\,\mu g\,C\,m^{-2}\,h^{-1}$ (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 CH₄ uptakes $(-75.4 \sim -158.4 \, \mu g \, C \, m^{-2} \, h^{-1})$ coincided with a period of elevated soil temperatures $(27.6 \sim 29.0 \, ^{\circ} C)$, optimum soil moisture (WFPS: $30.3 \sim 46.6 \, \%$) and groundwater table $(-0.8 \sim -1.0 \, m)$.

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 $5.57\pm0.68\,\mathrm{kg}\,\mathrm{N}\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$, 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 and Table 3). The slopes of

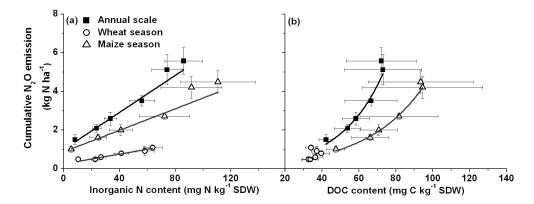


Fig. 6. Relationship between cumulative N₂O emissions and average soil contents of inorganic nitrogen (N) and dissolved organic carbon (DOC). SDW: soil dry weight. Error bars for vertical and horizontal ordinates represent standard error of the treatment averages and seasonal or annual averages, respectively.

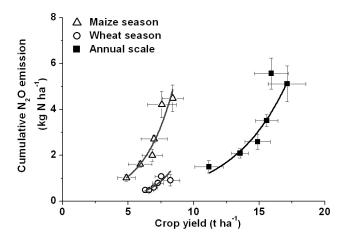


Fig. 7. Relationship between cumulative N₂O emissions and crop yield. Error bars represent standard error of the treatment averages.

linearly fitting curves (equal to the N_2O emission factors) were 0.0017 ± 0.0002 $(0.17 \pm 0.02\%)$, 0.0073 ± 0.0005 $(0.73 \pm 0.05\%)$ and 0.0049 ± 0.0002 $(0.49 \pm 0.02\%)$ for the wheat season, maize season and annual scale, respectively (Table 3).

The annual cumulative CH₄ uptakes for N0, N135, N270, N430, N650 and N850 were $-1.49\pm0.02,$ $-1.59\pm0.04,$ $-1.56\pm0.04,$ $-1.51\pm0.09,$ -1.69 ± 0.10 and -1.79 ± 0.07 kg C ha $^{-1}$, respectively (Table 2). The extremely high fertilizer rates $(350\sim450$ kg N ha $^{-1})$ in the maize season tended to increase the cumulative CH₄ uptake as compared with the low fertilizer rates $(0\sim250$ kg N ha $^{-1})$. The increased fertilizer rate $(0\sim400$ kg Nvha $^{-1})$ had no detectable effects on the cumulative CH₄ uptake in the wheat season (Figs. 4b, 5b and Table 3).

The combined CO_2 -equivelent emissions of N_2O and CH_4 on a 100-yr time horizon were estimated to be 650.9 ± 122.3 , 925.0 ± 95.3 , 1161.8 ± 156.1 ,

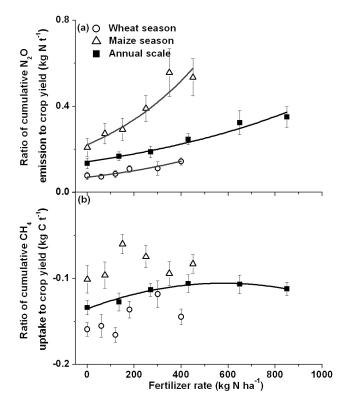


Fig. 8. Ratio of cumulative N_2O emissions and CH_4 uptake to crop yield across the nitrogen addition gradient. Error bars represent standard error of the treatment averages.

 1593.0 ± 122.7 , 2341.7 ± 365.4 and 2549.3 ± 318.0 kg CO₂ ha⁻¹ yr⁻¹, respectively, for N0, N135, N270, N430, N650 and N850. The increase of fertilizer rate also linearly enhanced the combined CO₂-equivelent emissions of N₂O and CH₄ (p < 0.01, Fig. 5c and Table 3).

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, Figs. 6a, b and Table 3). 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 (Figs. 6a, b and Table 3).

The exponential stimulation of N_2O emissions in the wheat and maize seasons followed the increase of crop yield induced by the increased fertilizer rate (Fig. 7 and Table 3). The ratios of cumulative N_2O emissions to maize and wheat yields were exponentially correlated with the fertilizer rate (Fig. 8a and Table 3). 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 ($\leq 135 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) resulted in relatively high cumulative CH_4 uptake per unit of crop yield in the wheat-maize rotation field (Fig. 8b and Table 3).

4 Discussion

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

Both the linear and nonlinear responses of cumulative N₂O 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 N₂O emissions to fertilizer rates could be well described by linear models for the wheat season, maize season and annual scale. The N₂O emissions were only a minor loss pathway of the applied fertilizer nitrogen in the wheat-maize rotation field $(0.49 \pm 0.02\%)$ of applied nitrogen in the annual scale). Ju et al. (2009) also reported very low averaged loss rates of N₂O emission to fertilizer nitrogen (0.12 and 0.23 % in the wheat and maize seasons) in the wheat-maize rotation fields on the North China Plain. They explained the low N2O loss rates by the low available carbon sources (organic matter: $1.0 \sim 1.5 \%$) and soil WFPS for denitrification in most semi-humid upland soils of the North China Plain. The low availability of soil organic carbon $(12.4 \sim 13.9 \,\mathrm{g\,kg^{-1}})$ hindered the transformation of NO₃⁻ to N₂O by denitrification and therefore caused the NO₃⁻ accumulation in the soil when high fertilizer nitrogen was applied (Wan et al., 2009; Ju et al., 2009, 2011). Finally, the accumulated NO₃ was leached out of the root zone due to the irrigation and concentrated rainfall in summer (Zhao et al., 2006; Ju et al., 2009). It's very likely that the denitrification process was also limited by the low availability of soil organic carbon $(11.3 \pm 0.6 \,\mathrm{g \, kg^{-1}})$ in our wheat-maize rotation field. Many studies have proved that under the native carbon condition of agricultural soils in northern China, the contribution of denitrification to N₂O emission was very limited (Wan et al., 2009; Ju et al., 2009; 2011; Cui et al., 2012). Therefore, the loss rates of N₂O emission to fertilizer nitrogen were relatively low and the increase of fertilizer rate did not exponentially stimulate the N₂O emission caused by the limited contribution of denitrification for most upland agricultural soils with low availability of soil organic carbon in northern China. However, due to the NO₃ accumulation in the soil arose by the lack of decomposable carbon and the extensive application of irrigation following fertilization, the NO₃⁻ leaching may be primarily stimulated when high fertilizer nitrogen was applied.

Aronson and Helliker (2010) performed a meta-analysis on the published CH₄ fluxes in the nitrogen amended and control plots. The results indicated that low fertilizer rates $(<100 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1})$ could stimulate CH₄ 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 CH₄ uptake by methanotrophic bacteria. The mechanism is believed to be responsible for the inhibition of CH₄ uptake in the soil exposed to high concentrations of available nitrogen. However, in our study, increased fertilizer rates ($0 \sim 400 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1}$) had no detectable effects on the cumulative CH₄ uptake by the soil in the wheat season. High fertilizer rates ($>350 \text{ kg N ha}^{-1}$) tended to enhance the cumulative CH₄ uptake by the soil in the maize season. The increased CH₄ consumption by ammonia oxidizers, changes in the community composition of methanotrophic bacteria, general improvement of nutrient availability and enhanced CH₄ diffusivity through plants may have been responsible for the enhanced effects (Bodelier and Laanbroek, 2004). The enhanced effects of high fertilizer rates on CH₄ 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 CH₄. We did observe weak CH₄ emissions ($<25.1 \,\mu g \, C \, m^{-2} \, h^{-1}$) after the continuous precipitation and irrigation events in the wheat-maize rotation field. The methanotrophic bacteria can benefit from the increased CH₄ 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 CH₄ consumption. Furthermore, after the irrigation and continuous precipitation the soil moisture for the treatments with high fertilizer rates decreased faster due to the possibly higher evapotranspiration caused by the higher aboveground biomass as compared with the low fertilizer rate treatments. It means that the CH₄ uptake could rapidly recover for the treatments with high fertilizer rate. Therefore, we observed delayed effects and enhanced effects of high fertilizer rates on atmospheric CH₄ uptake after the irrigation and continuous precipitation in the maize season in the wheat-maize rotation field.

4.2 Direct N₂O 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-N_2O}) for synthetic 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 $15 \sim 20 \, \mathrm{yr}$, especially in developing countries, which has significantly influenced the magnitude of EF_{F-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 N₂O 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 ($0 \sim 850 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$) covered the spatial and temporal variability of fertilizer rate in the region. Because the relationship between the cumulative N₂O emissions and fertilizer rates could be described well by linear models, the calculated N2O emission factors were constant for different nitrogen application rates $(0.17 \pm 0.02 \%)$ for the wheat season, 0.73 ± 0.05 % for the maize season and 0.49 ± 0.02 % for the annual scale). The estimated EF_{F-N₂O} of $0.49 \pm 0.02\%$ in the rotation year of 2009–2010 was slightly lower than the EF_{F-N_2O} of 0.67 \pm 0.23 % in the rotation year of 2008–2009 for the same experimental field (Liu et al., 2011). The direct N₂O emission factors for the wheatmaize rotation field in both rotation years were much lower than the global default EF_{F-N_2O} of croplands (1.25 \pm 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-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-N2O} (Ju et al., 2009, 2011).

4.3 Recommended rate of nitrogen fertilizer application

The optimized nutrient management should reduce nitrogen fertilizer input where fertilizer nitrogen has been overused, improve nitrogen use efficiency, and finally maintain soil nitrogen balance between inputs and outputs. In our wheat-maize rotation field, the estimated nitrogen inputs by irrigation (about 4 kg N ha⁻¹ yr⁻¹) and deposition (about $89 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, Ju et al., 2009; wet deposition: $30 \sim 43 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, this study and Liu et al., 2011) were approximately $93 \pm 11 \text{ kg N ha}^{-1} \text{ yr}^{-1}$. Applying the ratios of nitrogen losses reported by Ju et al. (2009), the processes of ammonia volatilization (approximately $60 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ 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) lost approximately $86 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$. The calculation of soil nitrogen balance between inputs (fertilization, irrigation and deposition) and outputs (grain, ammonia volatilization, nitrate leaching, gaseous nitrogen emissions by microbial denitrification) showed that only N270 could achieve a slightly positive soil nitrogen balance (gain $33 \pm 16 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$; fertilizer input: $270 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1}$ yr^{-1} ; grain output: $243 \pm 13 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ see Table 1). The slightly positive soil nitrogen balance means that the current crop yield $(7.0 \pm 0.3 \text{ and } 6.8 \pm 0.8 \text{ t ha}^{-1} \text{ for wheat})$ and maize) might be sustainable and the negative environmental effects could be minimized for N270. The treatments with higher fertilizer rates could obtain a small increase in the crop yield, but sharply stimulated the N₂O emission. We can see if the fertilizer rate increased from 270 to 430, 650 and $850 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$, the crop yield only increased 3.3%, 14.5% and 15.4% (0.5, 2.0 and $2.1 \, \text{tha}^{-1} \, \text{yr}^{-1}$) whereas the cumulative N₂O emissions increased 35.4%, 97.5 % and 114.9 % (0.9, 2.5 and $3.0 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1} \,\mathrm{yr}^{-1}$). Accordingly, we recommend 270 kg N ha⁻¹ yr⁻¹ as the optimized fertilizer rate for the local wheat-maize rotation fields to keep the current crop yield ($\sim 13.8 \,\mathrm{kg}\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$), to maintain the soil nitrogen balance (gain \sim 33 \pm 16 kg N ha⁻¹ yr^{-1}) and to reduce the N₂O emissions to the atmosphere $(\sim 2.59 \pm 0.33 \text{ kg N ha}^{-1} \text{ yr}^{-1})$. Then the fertilizer rates for local farmers' use $(430 \sim 470 \text{ kg N ha}^{-1} \text{ yr}^{-1})$, Liu et al., 2011) can be reduced by 37 % \sim 43 %.

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^- 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 suggest that the fertilizer rates of farmers' practice (430 to $670 \, \mathrm{kg} \, \mathrm{N} \, \mathrm{ha}^{-1} \, \mathrm{yr}^{-1}$) can be decreased by 30 to $60 \, \%$ to reduce the nitrogen losses to the environment and to maintain the current crop yield ($5.8 \sim 7.7 \, \mathrm{tha}^{-1}$ for wheat and $7.1 \sim 8.9 \, \mathrm{tha}^{-1}$ for maize). The reported optimum fertilizer rates for the wheat-maize rotation fields in northern

China vary 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 are within these ranges.

5 Conclusions

We investigated the crop yield, N₂O and CH₄ fluxes for the treatments of six fertilizer 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 responses of cumulative N₂O emissions to fertilizer rates could be described well by linear models for the wheat season, maize season and annual scale. The calculated N₂O emission factors (0.49 \pm 0.02 % for the annual scale) were adaptive to the different fertilizer rates in the wheat-maize rotation field. Increased fertilizer rates had no detectable effects on the cumulative CH₄ uptake by the soil in the wheat season whereas very high fertilizer rates ($\geq 350 \,\mathrm{kg} \,\mathrm{N} \,\mathrm{ha}^{-1}$) tended to enhance the CH₄ uptake in the maize season. The increase of crop yield induced by the increased fertilizer rates accompanied the exponential stimulation of N₂O emissions. We recommend the fertilizer rates for local farmers' use $(430 \sim 470 \text{ kg N ha}^{-1} \text{ yr}^{-1})$ to be reduced to 270 kg N ha^{-1} yr^{-1} to keep the current crop yield ($\sim 13.8 \, \text{t ha}^{-1} \, \text{yr}^{-1}$), to maintain the soil nitrogen balance, 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 fields.

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