

Nitrous oxide emissions from maize-wheat field

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Nitrous oxide emissions from maize-wheat field during four successive years in the North China Plain

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Abstract

Agricultural soil with fertilization is a main anthropogenic source for atmospheric N_2O . N_2O fluxes from a maize-wheat field in the North China Plain (NCP) were investigated for four successive years using static chamber method. The annual N_2O fluxes from control (without fertilization) and fertilization plots were 1.5 ± 0.2 and $9.4 \pm 1.7 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2008–2009, 2.0 ± 0.01 and $4.0 \pm 0.03 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2009–2010, 1.3 ± 0.02 and $5.0 \pm 0.3 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2010–2011, and 2.7 ± 0.6 and $12.5 \pm 0.1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2011–2012, respectively. Fertilizer-induced emission factors (EFs) in the corresponding years were 2.4, 0.60, 1.1 and 2.9%, respectively. Significant linear correlation between fertilized-induced N_2O emission (Y , $\text{kg N ha}^{-1} \text{ yr}^{-1}$) and rainfall 4 day before and 10 days after fertilization (X , mm) was found as $Y = 0.04767X - 1.06453$ ($N = 4$, $R^2 = 0.99241$, $P = 0.00253$). Therefore, the remarkable interannual variations of N_2O emissions and the EFs from the agricultural field were mainly ascribed to the rainfall. The total N_2O emission from the agricultural field in the NCP was estimated to be 144 Gg N yr^{-1} based on the average flux derived from the measurements of four years, and the fertilizer-induced N_2O emission accounted for about 76% (110 Gg N yr^{-1}) of total emission.

1 Introduction

Emissions of nitrous oxide (N_2O) to the atmosphere have attracted much attention because of its significance for greenhouse effect and depletion of stratospheric ozone (Crutzen, 1970; Bolle et al., 1986). Agricultural soil has been recognized as a main source of anthropogenic N_2O emissions to the atmosphere (Khalil et al., 2006) and contributes about 65% of total anthropogenic N_2O emission (Smith, 1997). It is well known that N_2O is a by-product in microbial nitrification and an intermediate in denitrification process (Firestone and Davidson, 1989). N_2O emissions from soils are strongly affected by many factors, e.g. soil temperature and moisture, soil aeration status and

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carbon availability (Smith et al., 2003; Ruser et al., 2006), crop type and residue management (Raich and Tufekcioglu, 2000; Huang et al., 2004; Chen et al., 2008), and the management of N fertilizer (Hao et al., 2001; Bouwman et al., 2002). Among the various influence factors, fertilization, soil temperature and moisture play important roles on N_2O emission. Fertilization directly provides substrate for soil nitrifying and denitrifying microbes, and soil temperature and moisture have major impacts on soil microorganisms (Smith et al., 2003). The microbial process generally increases exponentially with soil temperature when other factors are not limiting (Meixner and Yang, 2006). Soil water content plays important roles not only on the substrate supply for the microorganisms (Meixner and Yang, 2006) but also on gas diffusivity (Smith et al., 2003). Increasing soil moisture is conducive to produce anaerobic condition and thus promotes N_2O formation via denitrification (Dobbie and Smith, 2001). Large temporal-spatial variation of N_2O emission from agricultural fields could be expected due to the changes of the various influence factors, e.g. there are great uncertainties in N_2O emission from agricultural fields with the reported emission factors (EFs) of 0–7 % for mineral soils (Bouwman, 1996). Therefore, it is necessary to conduct long-term N_2O flux measurements from different agricultural field to reduce the uncertainty of N_2O estimation (Barton et al., 2008; Scheer et al., 2008).

North China Plain (NCP) is one of the greatest grain production areas in China. Maize and wheat, the main grain crops in this region, provide 39 % and 48 % of the total maize and wheat yields in China, respectively (Liu and Mu, 1993). The NCP has a cultivated land area of 17.95 million ha, which accounts for 18.6 % of the total agricultural area in China (Liu et al., 2001), and consumes about 30 % of the total national N-fertilizer (Zhang et al., 2004). As N-fertilizer is the necessary substrate for soil nitrification and denitrification, the huge amounts of N-fertilizer applications in this region can greatly stimulate N_2O emission. Therefore, N_2O emissions from the agricultural fields in the NCP have been investigated intensively (Zeng et al., 1995; Dong et al., 2000; Meng et al., 2005; Ding et al., 2007; Sun et al., 2008; Wang et al., 2008; Wang et al., 2009; Li et al., 2010; Cui et al., 2012; Cai et al., 2013). However, among the ten re-

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ports on N₂O emissions from the fields, nine studies conducted one year, and only the study of Cai et al. (2013) implemented the N₂O measurement for three years (2004–2007). According to these treatment-site-year data, large differences of N₂O emissions (ranging from 0.77 to 6.0 kg N ha⁻¹ yr⁻¹) and EFs (in the range of 0.10–1.4 %) from the agricultural fields in the NCP were obtained.

In this study, the N₂O flux from maize-wheat rotation system in the NCP was investigated from 2008 to 2012. The objectives of this study were: (1) to understand the interannual variation characters of N₂O emission, (2) to determine the key influence factors on N₂O emission and (3) to assess the total N₂O emission from the maize-wheat field in the NCP.

2 Materials and methods

2.1 Field experiment

This study was conducted in a summer maize (*Zea mays* L.) and winter wheat (*Triticum aestivum* L.) rotation system in Wangdu County (38°71′ N, 115°15′ E), Baoding City, Hebei Province, China. The detail information about the experiment field had been mentioned in our previous papers (Zhang et al., 2011, 2012).

The field experiment was conducted with two different treatments: control plot (CK, without fertilization) and chemical N fertilizer plot (NP). Only with the exception of fertilization, the two plots were identically managed. Each plot (6.5 × 3.5 m²) was separated by a 1.2 m broad zone to prevent nutrient transfer between treatments. Maize and wheat were planted in June and October each year, respectively, and the field was tilled before wheat sowing. Field managements including fertilization, irrigation, herbicide and pesticide applications strictly followed the cultivating manner of local farmers. The detail information about fertilizer management is listed in Table 1.

2.2 N₂O fluxes measurement

N₂O fluxes were investigated in the summer maize-winter wheat field from June in 2008 to October in 2012. Static chambers (60 × 60 × 90 cm³) were adopted to monitor N₂O fluxes. Three stainless steel pedestals were inserted 10 cm into the soils of each plot during the whole growing season. Four maize seeds (in June) and about 280 wheat seeds (in October) were kept in each pedestal, respectively. N₂O flux was measured every day with duration of at least 10 days after fertilization, then once or twice weekly during other periods of crops' growing seasons. On each sampling day, N₂O flux was measured at 9.30 a.m. (Beijing time).

N₂O concentrations were determined using a gas chromatography (Model SP3410, Beijing Analytical Instrument Factory) equipped with ⁶³N electron capture detector (Zhang et al., 2011, 2012). The N₂O flux (F , ngNm⁻²s⁻¹) was calculated by the following equation:

$$F = H \times \frac{\Delta C}{\Delta t} \times \frac{P}{RT} \times M_N \times 10^3. \quad (1)$$

where H is the chamber headspace height (m), $\Delta C/\Delta t$ is the slope (ppbvs⁻¹) of the linear regression of N₂O concentration in the chamber with time ($R^2 > 0.85$), P is the atmospheric pressure (atm) measured in the field, R is the gas constant (0.082 atmLK⁻¹mol⁻¹), T is the ambient air temperature (K) and M_N is the molecular weight of N₂O-N (28 gmol⁻¹).

2.3 Measurement of soil characteristics

Four soil samples in each plot were collected from 0–10 cm soil layer using a stainless steel soil sampler and were mixed carefully for the analysis of soil mineral N (NH₄⁺-N and NO₃⁻-N concentrations). Soil water filled pore space (WFPS) was determined at 5 cm depth by a ring sampler (100 cm³) (Zhang et al., 2011, 2012). Soil temperature was recorded on each gas-sampling day at a depth of 10 cm, while only the

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soil temperatures in CK plot were recorded in 2008. The data of precipitation were from <http://www.wunderground.com>.

2.4 Date calculation and statistical analysis

The statistical analysis was conducted by Origin 8.0 (Origin Lab Corporation, USA) and SPSS 13.0 software (SPSS Inc., Chicago, USA). Prior to analysis, normal distributions of N₂O fluxes and driving factors were tested using the Shapiro–Wilk test and data were log-transformed as needed to normalize the distributions. Paired-samples *T* test was adopted to analyze the difference between CK and NP treatments during the no fertilization periods. Stepwise linear regression analysis was performed to examine the relationships between N₂O fluxes and important driving factors, and only the regression equations that have statistical significance are listed in this study. Significance of all tests was accepted at $P < 0.05$.

The N₂O fluxes presented in the figures are the arithmetic means of the replications in each treatment. The cumulative N₂O emission from each treatment was estimated by linear interpolations between the sampling days. The EFs during the investigation periods were calculated as the difference between the cumulative N₂O-N emission in the fertilized plot and control plot divided by the amount of N fertilizer applied.

3 Results

3.1 Environmental variables

The variations of soil moisture were mainly regulated by precipitation and irrigation. Generally, soil moisture would increase quickly after irrigation (WFPS > 60%), and it could reach 80% or above when precipitation happened just after irrigation (Fig. 1a–d). The annual precipitation was 352, 356, 306 and 383 mm during the 2008–2009, 2009–2010, 2010–2011 and 2011–2012 maize-wheat seasons, respectively, and the precipitation in the maize season accounted for 75%, 62%, 64% and 79% of the

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total amount in each year, respectively. The mean soil moistures (WFPS) in the CK and NP plots were 57 % and 65 %, 48 % and 64 %, 55 % and 55 %, 68 % and 69 % in the 2008–2009, 2009–2010, 2010–2011 and 2011–2012 maize-wheat rotation years, respectively. The average soil temperatures of the CK and NP plots were 26.7, 22.5, 26.4 and 25.5 °C in the 2008, 2009, 2010 and 2011 maize growing seasons, and were 12.9, 11.0, 11.0 and 9.3 °C in the corresponding wheat seasons, respectively.

3.2 N₂O fluxes and key influence factors

The temporal variations of N₂O fluxes from the CK and NP plots during the four years are illustrated in Fig. 2. N₂O emissions from the CK plot were in the range of –37–70 ngNm⁻²s⁻¹, and obvious emission pulses occasionally occur after irrigation and rainfall events. As for the NP plot, the relatively high N₂O emissions (75–624 ngNm⁻²s⁻¹) usually occurred after fertilization, and the N₂O emission was from –19 to 33 ngNm⁻²s⁻¹ during the periods of pre- and post-fertilizer application. Negative N₂O fluxes (uptake, i.e. fluxes from the atmosphere to the soil) were occasionally observed in the CK and NP plots in this study, which accounted for 4–10 % of total investigation data in each maize-wheat season. The lowest detectable flux of the GC-ECD is 0.57 ngNm⁻²s⁻¹ in this study, and thus the extremely low N₂O uptakes are probably caused by the fluctuations of instrument. Nevertheless, the larger N₂O uptakes (–7 to –37 ngNm⁻²s⁻¹ in the CK plot; –7 to –19 ngNm⁻²s⁻¹ in the NP plot) can be ascribed to denitrification and nitrifier denitrification by reduction of N₂O to N₂ (Chapuis-lardy et al., 2007). Yamulki et al. (1995) and Mahmood et al. (1998) also reported evident negative N₂O fluxes from agricultural fields.

As shown in Fig. 2, N₂O emission peaks induced by fertilization usually occurred at the 1st–5th day after fertilization following irrigation in each growing season, while they delayed 1–2 days when rainfall events occurred just after fertilizations, e.g. on 21 August 2008, 5 July and 5 August 2009 (Fig. 2a and b). Generally, the N₂O peaks only lasted for one day and then decreased quickly, while the high N₂O emissions

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and the similar conclusion has been drawn by other studies (Wang et al., 2005; Rowlings et al., 2012).

3.3 Cumulative N₂O emissions and emission factors

The cumulative N₂O emissions and EFs are listed in Table 3. The lowest emissions always occurred in the CK plot, with the 4 yr mean fluxes of 0.6 kg N ha⁻¹ in the maize season, 1.3 kg N ha⁻¹ in the wheat season and 1.9 kg N ha⁻¹ in the whole year. The annual cumulative N₂O emissions from the NP plot in 2009–2010 and 2010–2011 were close, and extremely high N₂O emissions were observed in 2008–2009 and 2011–2012 maize-wheat seasons. Mean cumulative N₂O emissions from the NP plot in the maize, wheat growing seasons and the whole year were 4.4, 3.3 and 7.7 kg N ha⁻¹, with the variation coefficients of 46, 90 and 51 %, respectively.

The annual EFs were 2.4 %, 0.6 %, 1.1 % and 2.9 % in 2008–2012 maize-wheat seasons, respectively (Table 3). The mean 4 yr EF in the maize season (2.2 %) was 1.8 times higher than that in the wheat season.

4 Discussion

4.1 Interannual variation of N₂O emission

The above results well revealed evident interannual variation of N₂O emissions from the agricultural field during the four successive years. Considering the nearly identical N-fertilization rates and similar irrigation operations in each year, the interannual variation of N₂O emissions was mainly ascribed to the changes of meteorological condition that affected the soil temperature and moisture. The annual cumulative N₂O emissions (F_1 , kg N ha⁻¹) from the CK plot significantly correlated with the annual total rainfall (X_1 , mm), and the relationship fitted the following equation:

$$F_1 = 5.6 \times 10^{-10} X_1^{3.7}, \quad N = 4, \quad R^2 = 0.754, \quad P = 0.013. \quad (2)$$

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It indicated that rainfall was a dominant factor for controlling N_2O emission from the agricultural field without fertilization. As for the NP plot, there was no evident correlation between the annual cumulative N_2O emissions and total rainfall. However, there was a significant linear correlation ($N = 4$, $R^2 = 0.99241$, $P = 0.00253$) between the annual fertilizer-induced N_2O emissions (F_2 , kgNha^{-1}) during the periods of 10 days after fertilization and the amounts of rainfall (X_2 , mm) 4 days before- and 10 days after each fertilization in each year. The equation could be expressed as:

$$F_2 = 0.04767X_2 - 1.06453. \quad (3)$$

Because the cumulative N_2O emission from the NP plot during the periods of 10 days after fertilization accounted for $\sim 50\%$ of the total cumulative N_2O emissions, the rainfall events far from fertilization events only made modest contribution to the total cumulative N_2O emissions. The rainfall events just before and after fertilization might favor fostering the community of microorganism and promoting N_2O formation. Therefore, it is easy to understand why the strong correlation only limited between the yearly fertilizer-induced N_2O emissions during the periods of 10 days after fertilizations and the amounts of rainfall around fertilization events in each year.

There were no significant differences between the CK and NP treatments during the no fertilization periods in the four years (T test, $P = 0.056\text{--}0.177$) and the yearly management (fertilization rate and irrigation) of the agricultural field was almost the same in recent years. Only based on the rainfall, the correlations between the N_2O emissions and the rainfall could be useful to estimate the cumulative N_2O emissions (F , kgNha^{-1}) from the agricultural field:

$$F = 5.6 \times 10^{-10}X_1^{3.7} + 0.04767X_2 - 1.06453. \quad (4)$$

It should be mentioned that the above estimation could be only applied to limit area where the rainfall is nearly identical and fertilization rate is similar with this study. Because the cumulative N_2O emissions usually linearly correlate with fertilization rates

(Henault et al., 1998; De Klein et al., 2006; Halvorson et al., 2008), the above experiential algorithm for estimating the annual cumulative N₂O emissions (Y , kgNha⁻¹) from agricultural fields could be expressed in more general form:

$$Y = AX_1^n + F/BX_2 - C, \quad (5)$$

where n , A , B and C are constants which can be derived from the correlation of field measurements, and F is the application rate of N-fertilizer. To verify the applicability of the above algorithm, more field studies in various agricultural fields are needed. If the applicability of the above algorithm was verified, the global annual cumulative N₂O emissions from agricultural fields could be easily estimated just based on fertilization rates and rainfall in different regions.

4.2 Comparison with previous studies and assessing the total N₂O emission in the NCP

The results of studies from maize-wheat fields in the NCP are shown in Table 4. It is evident that there are very large temporal-spatial variations of the cumulative N₂O emissions and EFs reported in the NCP. With only the exception of the data in 2009–2011 maize-wheat seasons, the cumulative N₂O emissions from the NP plot in this study were 33–108 % greater than the upper limit value reported in the literatures. The EF value of 0.60 % in 2009–2010 was in good agreement with the values reported by Dong et al. (2000) and Ding et al. (2007), and of 1.1 % in 2010–2011 was in line with the values reported by Li et al. (2010), Sun et al. (2008) and Cai et al. (2013). The EFs from the NP plot in 2008–2009 and 2011–2012 were two times greater than the upper limit value reported in the NCP, but were still within the uncertainty range recommended by the IPCC (0.3–3 %, De Klein et al., 2006).

To check the possible influence of the soils from different areas in the NCP on N₂O emission, soil samples were collected from four sampling sites (Fengqiu, Luancheng, Yucheng and Beijing) where N₂O emissions have been investigated. N₂O emissions from the four fields were simultaneously measured under the same fertilization and

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Table 1. The amount, date and type of nitrogen fertilizer application in the experiment field.

Year	Fertilization timing		Fertilizer type		Fertilizer rate (kgNha ⁻¹)		
	B ^a	S ^b	B ^a	S ^b	B ^a	S ^b	T ^c
Maize							
2008	25 Jun 2008	16 Aug 2008	NPK ^d	NK ^d	89	83	172
2009	29 Jun 2009	1 Aug 2009	NPK ^d	Urea	99	69	168
2010	30 Jun 2010	6 Aug 2010	NPK ^d	NK ^d	107	69	176
2011	30 Jun 2011	8 Aug 2011	NPK ^d	NK ^d	107	69	176
Wheat							
2008–2009	18 Oct 2008	4 Apr 2009	NPK ^d	Urea	75	88	163
2009–2010	13 Oct 2009	20 Apr 2010	NPK ^d	NK ^d	60	105	165
2010–2011	17 Oct 2010	14 Apr 2011	NPK ^d	NS ^d	60	105	165
2011–2012	19 Oct 2011	18 Apr 2012	NPK ^d	Urea	60	105	165

^a Basal fertilizer; ^b Supplemental fertilizer; ^c Total fertilization rate; ^d Compound fertilizers contained nitrogen (N), phosphorus (P), potassium (K) and sulfur (S).

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Table 2. Regression analysis between N_2O flux and its control factors in the maize-wheat field.

Treatment	Factors	B^a	R^2	Equation
2009–2010 maize-wheat season				
CK	WFPS	0.021	0.306	$\lg\text{N}_2\text{O} = 0.021\text{WFPS} + 0.052\text{ST} - 0.935$
	ST^b	0.052	0.516	$(N = 17, R = 0.817, P < 0.001)$
NP	WFPS	0.012	0.184	$\lg\text{N}_2\text{O} = 0.012\text{WFPS} + 0.334\lg\text{NH}_4^+\text{-N} + 0.488$
	$\lg\text{NH}_4^+\text{-N}$	0.334	0.287	$(N = 20, R = 0.684, P = 0.005)$
2010–2011 maize-wheat season				
NP	WFPS	0.018	0.225	$\lg\text{N}_2\text{O} = 0.018\text{WFPS} + 0.041\text{ST} + 0.011\text{NO}_3^-\text{-N}$
	ST^b	0.041	0.270	$+ 0.599\lg\text{NH}_4^+\text{-N} - 1.412$
	$\text{NO}_3^-\text{-N}$	0.011	0.067	$(N = 30, R = 0.612, P = 0.016)$
	$\lg\text{NH}_4^+\text{-N}$	0.599	0.118	
2011–2012 maize-wheat season				
NP	$\lg\text{NH}_4^+\text{-N}$	0.767	0.196	$\lg\text{N}_2\text{O} = 0.767\lg\text{NH}_4^+\text{-N} + 0.871$ $(N = 28, R = 0.475, P = 0.011)$

^a Unstandardized coefficient; ^b Soil temperature.

Table 3. N₂O fluxes (mean ± SE) and EFs from different treatments in different years.

Year	Period	Treatment	N ₂ O cumulative fluxes (kg N ha ⁻¹)	EF (%)
2008–2009 maize-wheat season	Maize	CK	0.6 ± 0.2	–
		NP	7.2 ± 1.2	3.8
	Wheat	CK	0.9 ± 0.001	–
		NP	2.2 ± 0.5	0.80
	Annual	CK	1.5 ± 0.2	–
		NP	9.4 ± 1.7	2.4
2009–2010 maize-wheat season	Maize	CK	0.9 ± 0.02	–
		NP	2.8 ± 0.02	1.1
	Wheat	CK	1.1 ± 0.01	–
		NP	1.3 ± 0.03	0.12
	Annual	CK	2.0 ± 0.01	–
		NP	4.0 ± 0.03	0.60
2010–2011 maize-wheat season	Maize	CK	0.4 ± 0.01	–
		NP	3.0 ± 0.1	1.5
	Wheat	CK	0.8 ± 0.03	–
		NP	2.0 ± 0.1	0.73
	Annual	CK	1.3 ± 0.02	–
		NP	5.0 ± 0.3	1.1
2011–2012 maize-wheat season	Maize	CK	0.5 ± 0.3	–
		NP	4.7 ± 0.3	2.4
	Wheat	CK	2.3 ± 0.3	–
		NP	7.8 ± 0.4	3.3
	Annual	CK	2.7 ± 0.6	–
		NP	12.5 ± 0.1	2.9

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**Table 4.** Summary of N₂O emissions from maize-wheat soils in the NCP.

Location	Total N (kg N ha ⁻¹ yr ⁻¹)	Accumulative fluxes (kg N ha ⁻¹ yr ⁻¹)	EF (%)	References
Wangdu, Hebei	335	9.4	2.4	This study, 2008–2009
	333	4.0	0.60	This study, 2009–2010
	341	5.0	1.1	This study, 2010–2011
	341	12.5	2.9	This study, 2011–2012
Quzhou, Heibei	270	4.9	0.96	Li et al. (2010)
	135	5.0	1.0	Wang et al. (2008)
	270	6.0	0.87	
Luancheng, Hebei	200	0.89	0.12	Wang et al. (2009)
	400	1.1	0.10	
	600	1.4	0.13	
	300	1.6	0.23	Zeng et al. (1995)
Yucheng, Shandong	420	2.9	0.67	Dong et al. (2000)
	312	4.4	1.4*	Sun et al. (2008)
Huantai, Shangdong	600	4.0	0.59	Cui et al. (2012)
Fengqiu, Henan	300	0.77	0.21	Meng et al. (2005)
	300	2.5	0.61	Ding et al. (2007)
	500	4.5	0.77	
	300	2.4	0.63	Cai et al. (2013)
	300	3.0	0.95	
	300	2.9	0.88	

* Background N₂O emission wasn't subtracted.

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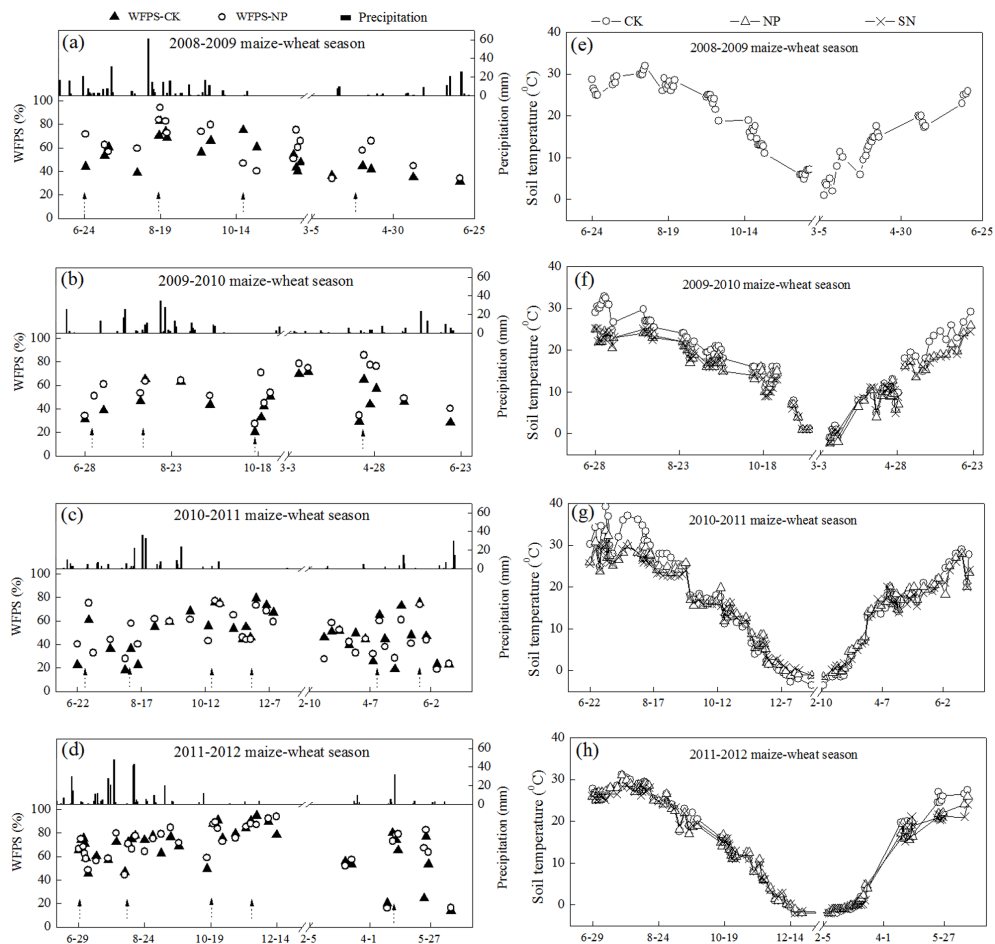


Fig. 1. Precipitation, soil WFPS (a, b, c, d) and soil temperatures (e, f, g, h) in the CK and NP plots during 2008–2012 maize-wheat seasons. Dash arrows show irrigation events.

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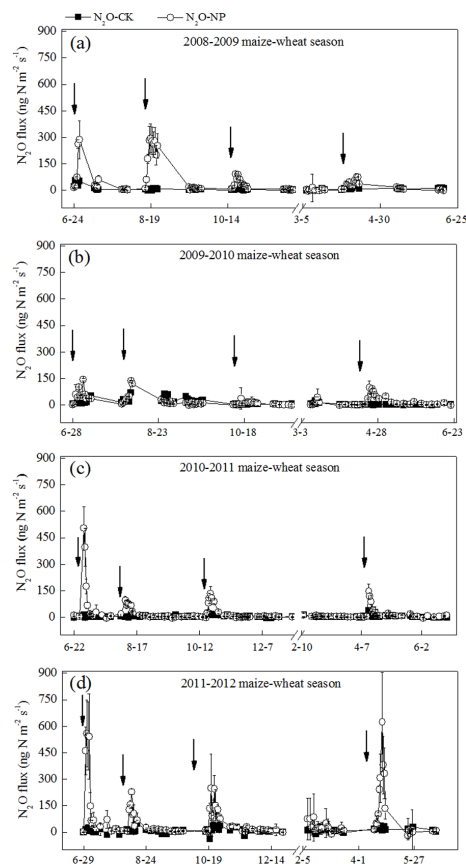


Fig. 2. N₂O emissions from the CK and NP plots during the N₂O measurement periods in 2008–2009 maize-wheat season (a), 2009–2010 maize-wheat season (b), 2010–2011 maize-wheat season (c) and 2011–2012 maize-wheat season (d). Arrows show fertilizer applications.

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