Biogeosciences, 11, 1717–1726, 2014
www.biogeosciences.net/11/1717/2014/
doi:10.5194/bg-11-1717-2014

Biogeosciences
Open Access

Nitrous oxide emissions from maize–wheat field during 4 successive years in the North China Plain

Y. Zhang, Y. Mu, Y. Zhou, J. Liu, and C. Zhang

Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

Correspondence to: Y. Mu (yjmu@rcees.ac.cn)

Received: 17 October 2013 – Published in Biogeosciences Discuss.: 26 November 2013
Revised: 17 February 2014 – Accepted: 18 February 2014 – Published: 1 April 2014

Abstract. Agricultural soil with fertilization is a main anthropogenic source for atmospheric nitrous oxide (N_2O). N_2O fluxes from a maize–wheat rotation field in the North China Plain (NCP) were investigated for 4 successive years using the static chamber method. The annual N_2O fluxes from the control (without fertilization) and fertilization plots were 1.5 ± 0.2 and 9.4 ± 1.7 kg N ha^{-1} yr^{-1} in 2008–2009, 2.0 ± 0.01 and 4.0 ± 0.03 kg N ha^{-1} yr^{-1} in 2009–2010, 1.3 ± 0.02 and 5.0 ± 0.3 kg N ha^{-1} yr^{-1} in 2010–2011, and 2.7 ± 0.6 and 12.5 ± 0.1 kg N ha^{-1} yr^{-1} in 2011–2012, respectively. Annual direct emission factors (EF_d's) in the corresponding years were 2.4 ± 0.5 %, 0.60 ± 0.01 %, 1.1 ± 0.09 % and 2.9 ± 0.2 %, respectively. Significant linear correlation between fertilized-induced N_2O emissions (Y, kg N ha^{-1}) during the periods of 10 days after fertilization and rainfall intensities from 4 days before to 10 days after fertilization (X, mm) in the 4 years was found as $Y = 0.048X - 1.1$ ($N = 4$, $R^2 = 0.99$, $P < 0.05$). Therefore, the remarkable interannual variations of N_2O emissions and the EF_d's were mainly ascribed to the rainfall.

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 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 nitrogen (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. For example, there are great uncertainties in N_2O emission from agricultural fields with the reported direct emission factors (EF_d's) of 0–7 % for mineral soils (Bouwman, 1996). Therefore, it is necessary to conduct long-term N_2O flux measurements different agricultural field to reduce the uncertainties 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).

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

Year	Fertilization timing		Fertilizer type		Fertilizer rate (kg N ha^{-1})		
	B	S	B	S	B	S	T
Maize							
2008	25 June 2008	16 August 2008	NPK	NK	89	83	172
2009	29 June 2009	1 August 2009	NPK	Urea	99	69	168
2010	30 June 2010	6 August 2010	NPK	NK	107	69	176
2011	30 June 2011	8 August 2011	NPK	NK	107	69	176
Wheat							
2008–2009	18 October 2008	4 April 2009	NPK	Urea	75	88	163
2009–2010	13October 2009	20 April 2010	NPK	NK	60	105	165
2010–2011	17 October 2010	14 April 2011	NPK	NS	60	105	165
2011–2012	19 October 2011	18 April 2012	NPK	Urea	60	105	165

B: basal fertilizer, S: supplemental fertilizer, T: total fertilization rate, NPK, NK, NS: compound fertilizers contained nitrogen (N), phosphorus (P), potassium (K) and sulfur (S).

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, 2013; Sun et al., 2008; Wang et al., 2008, 2009; Li et al., 2010; Cui et al., 2012; Cai et al., 2013; Hu et al., 2013; Shi et al., 2013; Yan et al., 2013). However, among those reports on N_2O emissions from the fields, many studies were conducted for 1 year, and only the studies of Cai et al. (2013), Hu et al. (2013) and Yan et al. (2013) implemented the N_2O measurement for more than 1 year. According to these treatment-site-year data, large differences of N_2O emissions (ranging from 0.77 to 6.0 kg N ha^{-1} yr^{-1}) and EF_d's (in the range of 0.10–1.0 %) from the agricultural fields in the NCP were obtained.

In this study, the N_2O flux from a 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 of N_2O emission and (2) to determine the key influence factors on N_2O emission.

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. Detailed information about the experiment field has been mentioned in our previous papers (Zhang et al., 2011, 2012).

The field experiment was conducted with two different treatments: control (CK, without fertilization) and chemical N-fertilizer (NP). Only with the exception of fertilization, the two plots were identically managed. Each plot ($6.5 \times 3.5\,m^2$) 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. Detailed information about fertilizer management is listed in Table 1.

2.2 N_2O fluxes measurement

N_2O fluxes were investigated in the summer maize–winter wheat field from June in 2008 to October in 2012. Static chambers ($60 \times 60 \times 90\,cm^3$) were adopted to monitor N_2O fluxes. Three stainless steel pedestals were inserted 10 cm into the soils of each plot during the whole growing season. The densities of sowing for maize and wheat in the pedestals were the same as the densities in the surrounding area. Four maize seeds (in June) and about 280 wheat seeds (in October) were kept in each pedestal, respectively. The top part of the maize plant above the chamber was cut off when its height exceeded 80 cm (after ∼ 40 days growing). N_2O fluxes were measured every day within duration of more than 10 days after fertilization (except the period of basal fertilizer application in 2008 maize season), then continuous sampling for 5–11 days monthly (2008–2010 maize–wheat rotations) or once to twice weekly (2010–2012 maize–wheat rotations) during other periods of crops' growing seasons excluding the winters (from December to February) in 2008–2010 maize–wheat rotations. On each sampling day, N_2O flux was measured at around 9.30 a.m. (Beijing time). Four gas samples were taken from the headspace by a sampling minipump (NMP 830 KNDC, Germany) to aluminum combined polyester gas sampling bags (200 mL, Delin, Dalian, China) at 10 min intervals after the chambers were deployed. The first sample was taken after 2 min of covering chambers.

N_2O concentrations were determined using a gas chromatograph (GC, Model SP3410, Beijing Analytical Instrument Factory) equipped with ^{63}N electron capture detector (ECD). An improved GC-ECD method was applied to measure N_2O concentration in this study (Zhang et al., 2013). High purity of N_2 (99.999 %) was used as carrier gas, and a makeup gas (979 ppmv CO_2 in N_2) was introduced into the downstream of the analytical column. The variation coefficient of our method for analyzing N_2O was less than 0.31 %. The negligible influence of CO_2 on N_2O measurement and the good linear correlation between the GC-ECD responses and N_2O concentrations were found by our improved GC-ECD method (Zhang et al., 2013). The N_2O flux (F, ng N m^{-2} s^{-1}) was calculated by the following equation:

$$F = H \times \frac{\Delta C}{\Delta t} \times \frac{P}{RT} \times M_{\mathrm{N}} \times 10^3, \quad (1)$$

where H is the chamber headspace height (m), C/t is the slope (ppbv s^{-1}) of the linear regression of N_2O 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 atm L K^{-1} mol^{-1}), T is the ambient air temperature (K) and M_{N} is the molecular weight of N_2O-N (28 g mol^{-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 (NH_4^+-N and NO_3^--N) concentrations by a colorimetric continuous flow analyzer (SANT++, Skalar Company, the Netherlands). The samples from 0 to 5 cm topsoil were determined gravimetrically by oven drying at 105 °C for 12 h and expressed as water-filled pore space (WFPS). Total porosity and WFPS were calculated based on water content, soil bulk density and a particle density of 2.65 g cm^{-3}. Soil temperature was recorded on each gas-sampling day at a depth of 10 cm, while only the soil temperatures in the CK treatment 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, data of N_2O fluxes and driving factors 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 periods of 4 years excluding the 10-day durations following each fertilization event. Stepwise linear regression analysis was performed to examine the relationships between N_2O 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 N_2O fluxes presented in the figures are the arithmetic means of the replications in each treatment. The cumulative N_2O emission from each treatment was estimated by linear interpolation between the sampling days. The EF_d's during the investigation periods were calculated as the difference between the cumulative N_2O-N emission from the fertilized and control plots divided by the amount of N-fertilizer applied. The standard error of the direct emission factor was estimated using the standard errors for the cumulative emissions from the fertilized and the unfertilized plots (Cui et al., 2012).

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 rotations, respectively, and the precipitation in the maize season accounted for 75, 62, 64 and 79 % of the total amount in each year, respectively. The mean soil moisture (WFPS) values 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 N_2O fluxes and key influence factors

The temporal variations of N_2O fluxes from the CK and NP plots during the 4 years are illustrated in Fig. 2. N_2O emissions from the CK treatment were in the range of −37–70 ng N m^{-2} s^{-1}, and obvious emission pulses occasionally occur after irrigation and rainfall events. As for the NP treatment, the relatively high N_2O emissions (75–624 ng N m^{-2} s^{-1}) usually occurred after fertilization, and the N_2O emission was from −19 to 33 ng N m^{-2} s^{-1} during the periods of pre- and post-fertilizer application. Negative N_2O 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 rotation. The N_2O concentrations were usually measured within 10 days, and the variation coefficient of N_2O concentrations in gas bags during

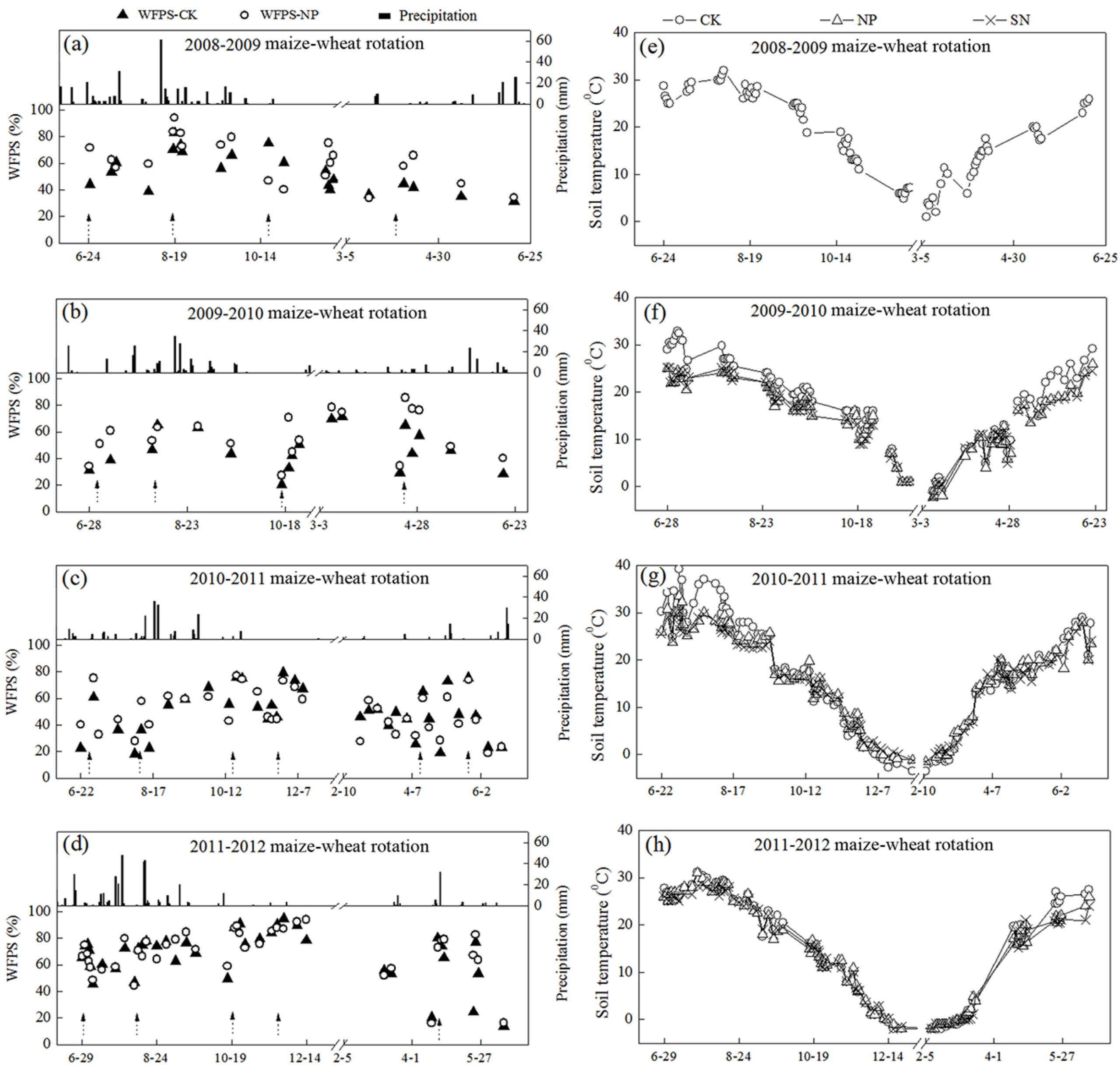

Fig. 1. Precipitation, soil WFPS **(a, b, c, d)** and soil temperatures **(e, f, g, h)** in the CK and NP treatments during 2008–2012 maize–wheat rotations. Dashed arrows show irrigation events.

storage and transportation was less than 1 %. Considering the flux detection limit was 1.6 ng N m^{-2} s^{-1} in this study, the negative fluxes close to the flux detection limit could be ascribed to the instable instrument signal, while the large uptakes (−37 ng N m^{-2} s^{-1} in the CK and −19 ng N m^{-2} s^{-1} in the NP treatments) were due to the denitrification and nitrifier denitrification (Chapuis-Lardy et al., 2007). Many researches also reported evident negative N_2O fluxes from agriculture fields (e.g., Yamulki et al., 1995; Mahmood et al., 1998; Cui et al., 2012; Yan et al., 2013).

As shown in Fig. 2, N_2O emission peaks induced by fertilization usually occurred at the first through fifth 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 N_2O peaks only lasted for 1 day and then decreased quickly, while the high N_2O emissions (about 550 ng N m^{-2} s^{-1}) sustained 3 days after basal fertilization following showers (from 1 July to 2 July) in 2011 maize season (Fig. 2d). Therefore, precipitation coincided with the fertilization would probably promote

Table 2. Regression analysis between N_2O flux (ng N m^{-2} s^{-1}) and its regulating factors in the maize–wheat field.

Treatment	Factors	*B*	R^2	Equation
				2009–2010 maize–wheat rotation
CK	WFPS	0.021	0.31	$\lg N_2O = 0.021\text{WFPS} + 0.052\text{ST} - 0.94$
	ST	0.052	0.52	$(N = 17, R = 0.82, P < 0.001)$
NP	WFPS	0.012	0.18	$\lg N_2O = 0.012\text{WFPS} + 0.33\lg NH_4^+\text{-N} + 0.49$
	$\lg NH_4^+$-N	0.33	0.29	$(N = 20, R = 0.684, P < 0.01)$
				2010–2011 maize–wheat rotation
NP	WFPS	0.018	0.23	$\lg N_2O = 0.018\text{WFPS} + 0.041\text{ST} +$
	ST	0.041	0.27	$0.011NO_3^-\text{-N} + 0.60\lg NH_4^+\text{-N} - 1.4$
	NO_3^--N	0.011	0.067	$(N = 30, R = 0.61, P < 0.05)$
	$\lg NH_4^+$-N	0.60	0.12	
				2011–2012 maize–wheat rotation
NP	$\lg NH_4^+$-N	0.77	0.20	$\lg N_2O = 0.77\lg NH_4^+\text{-N} + 0.87$
				$(N = 28, R = 0.48, P < 0.05)$

B: regression coefficient, ST: soil temperature (°C) at a depth of 10 cm; unit of mineral N (NO_3^--N and NH_4^+-N): mg kg^{-1} dry soil.

N_2O emission because of the substrate supplement and development of anaerobic soil condition.

The N_2O emission peaks from the NP treatment were 294, 142, 503 and 558 ng N m^{-2} s^{-1} in 2008, 2009, 2010 and 2011 maize seasons, respectively, and were 75, 100, 147 and 624 ng N m^{-2} s^{-1} in 2008–2009, 2009–2010, 2010–2011 and 2011–2012 wheat seasons, respectively. The N_2O emission peaks after basal or supplemental fertilizer application were usually higher during the maize seasons than during the wheat seasons, which might be due to the relatively low soil temperature in the wheat seasons (Fig. 1e–h). However, the maximal peak of N_2O emission (624 ng N m^{-2} s^{-1}) from the NP treatment among the 4 investigated years appeared in the 2012 wheat season after the supplemental fertilization, which was 4–8 times higher than those in other wheat seasons. During the period of N_2O peak emission from the NP treatment in the 2012 wheat season, the soil WFPS (78 %) was evidently higher than those in 2009 (66 %) and 2011 (60 %) wheat seasons (Fig. 1a, c and d). Although higher WFPS (82 %) was observed after the supplemental fertilizer application in the wheat season of 2010, the obvious low soil temperature (10 °C) compared with that (15.5 °C) in 2012 greatly restricted the activities of soil microorganisms (Meixner and Yang, 2006). Therefore, the higher N_2O emission in the wheat season of 2012 was due to the synergistic effect of appropriate soil temperature (15.5 °C) and WFPS (78 %), which could build the soil microenvironment in favor of denitrification, and thus promote the N_2O emission (Dobbie and Smith, 2001).

To elucidate the influence of various driving factors on N_2O emission, the regression analysis between N_2O fluxes and important driving factors was conducted as shown in Table 2. Evidently, N_2O emission positively correlated with soil mineral N, temperature and WFPS. Soil temperature and WFPS could explain 27–52 % and 18–31 % of the N_2O emission, respectively. However, not all factors displayed significant influences on the N_2O emission in each year and treatment, and a similar conclusion has been drawn by other studies (Wang et al., 2005; Rowlings et al., 2012).

3.3 Cumulative N_2O emissions and emission factors

The cumulative N_2O emissions and EF_d's are listed in Table 3. The lowest emissions always occurred in the CK treatment, with the 4-year 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 N_2O emissions from the NP treatment in the 2009–2010 and 2010–2011 were close, and extremely high N_2O emissions were observed in the 2008–2009 and 2011–2012 maize–wheat rotations. Mean cumulative N_2O emissions from the NP treatment 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 EF_d's were 2.4 ± 0.5 %, 0.6 ± 0.01 %, 1.1 ± 0.09 % and 2.9 ± 0.2 % in 2008–2012 maize–wheat rotations, respectively (Table 3). The mean 4-year EF_d in the maize season (2.2 %) was 1.8 times higher than that in the wheat season.

4 Discussion

4.1 Interannual variation of N_2O emission

The above results well revealed evident interannual variation of N_2O emissions from the agricultural field during the 4 successive years. Considering the nearly identical N-fertilization rates and similar irrigation operations in each year, the interannual variation of N_2O emissions was mainly ascribed to

Table 3. Cumulative nitrous oxide (N_2O) fluxes (mean ± SE) and direct emission factors (EF_d's, mean ± SE) from different treatments of the investigated rotations.

	Period	Treatment	Fluxes ($kg\,N\,ha^{-1}$)	EF_d (%)
2008–2009 rotation	Maize	CK	0.6 ± 0.2	–
		NP	7.2 ± 1.2	3.8 ± 0.7
	Wheat	CK	0.9 ± 0.001	–
		NP	2.2 ± 0.5	0.80 ± 0.31
	Annual	CK	1.5 ± 0.2	–
		NP	9.4 ± 1.7	2.4 ± 0.5
2009–2010 rotation	Maize	CK	0.9 ± 0.02	–
		NP	2.8 ± 0.02	1.1 ± 0.02
	Wheat	CK	1.1 ± 0.01	–
		NP	1.3 ± 0.03	0.12 ± 0.02
	Annual	CK	2.0 ± 0.01	–
		NP	4.0 ± 0.03	0.60 ± 0.01
2010–2011 rotation	Maize	CK	0.4 ± 0.01	–
		NP	3.0 ± 0.1	1.5 ± 0.06
	Wheat	CK	0.8 ± 0.03	–
		NP	2.0 ± 0.1	0.73 ± 0.06
	Annual	CK	1.3 ± 0.02	–
		NP	5.0 ± 0.3	1.1 ± 0.09
2011–2012 rotation	Maize	CK	0.5 ± 0.3	–
		NP	4.7 ± 0.3	2.4 ± 0.2
	Wheat	CK	2.3 ± 0.3	–
		NP	7.8 ± 0.4	3.3 ± 0.3
	Annual	CK	2.7 ± 0.6	–
		NP	12.5 ± 0.1	2.9 ± 0.2

the changes of meteorological condition that affected the soil temperature and moisture. The annual cumulative N_2O emissions from the CK treatment (F_1, $kg\,N\,ha^{-1}$) significantly correlated with the annual total rainfall intensities (X_1, mm), and the relationship fitted the following equation:

$$F_1 = 5.6 \times 10^{-10} X_1^{3.7}, N = 4, R^2 = 0.75, P < 0.05. \quad (2)$$

It indicated that rainfall was a dominant factor for controlling N_2O emission from the agricultural field without fertilization. As for the NP treatment, there was no evident correlation between the annual cumulative N_2O emissions and rainfall intensities. Significant linear correlations were only observed between the fertilizer-induced N_2O emissions (F, $kg\,N\,ha^{-1}$) during the periods of 10 days after fertilization and the amounts of rainfall (X, mm) from 4 days before to 10 days after fertilization in the maize, wheat seasons and whole year. The equations could be orderly expressed as

$$F_{\mathrm{maize}} = 0.035 X_{\mathrm{maize}} - 0.48, \mathrm{N} = 4, R^2 = 0.93, P < 0.05, \quad (3)$$

$$F_{\mathrm{wheat}} = 0.077 X_{\mathrm{wheat}} - 0.11, \mathrm{N} = 4, R^2 = 0.94, P < 0.05, \quad (4)$$

$$F_{\mathrm{year}} = 0.048 X_2 - 1.1, \mathrm{N} = 4, R^2 = 0.99, P < 0.05. \quad (5)$$

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

There was no significant difference between the CK and NP treatments during the periods of 4 years excluding the 10-day durations following each fertilization event (T test, $P > 0.05$), and the yearly management (fertilization rate and irrigation) of the agricultural field was almost the same in recent years. Therefore, only based on rainfall, the annual cumulative N_2O emissions (F_{annual}, $kg\,N\,ha^{-1}$) from the agriculture field could be obtained by integrating the Eqs. (2) and (5) as follows:

$$F_{\mathrm{annual}} = 5.6 \times 10^{-10} X_1^{3.7} + 0.048 X_2 - 1.1. \quad (6)$$

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

Table 4. Summary of N_2O emissions from maize–wheat soils in the NCP.

Location	Year	Total N ($kg\,N\,ha^{-1}\,yr^{-1}$)	Cumulative fluxes ($kg\,N\,ha^{-1}\,yr^{-1}$)	EF_d (%)	References
Wangdu, Hebei	2008–2009	335	9.4	2.4	This study
	2009–2010	333	4.0	0.60	This study
	2010–2011	341	5.0	1.1	This study
	2011–2012	341	12.5	2.9	This study
Quzhou, Hebei	2004–2005	135	5.0	1.0	Wang et al. (2008)
		270	6.0	0.87	
	2005–2006	270	4.9	0.96	Li et al. (2010)
	2009–2010	380	2.4	0.41	Hu et al. (2013)
	2010–2011	380	1.9	0.37	
Luancheng, Hebei	1992–1993	300	1.6	0.23	Zeng et al. (1995)
	2007–2008	200	0.89	0.12	Wang et al. (2009)
		400	1.1	0.10	
		600	1.4	0.13	
Huantai, Shandong	2008–2009	600	4.0	0.59	Cui et al. (2012)
	2008–2009	644	4.0	0.58	Yan et al. (2013)
	2009–2010	464	2.5	0.48	Shi et al. (2013)
	2011–2012	647	5.3	0.75	
		467	4.8	0.96	
		600	2.2	0.21	
		218	1.8	0.37	
Yucheng, Shandong	1995–1996	420	2.9	0.67	Dong et al. (2000)
	2004–2005	312	4.4	–	Sun et al. (2008)
Fengqiu, Henan	2002–2003	300	0.77	0.21	Meng et al. (2005)
	2004–2005	300	2.5	0.61	Ding et al. (2007)
		500	4.5	0.77	
	2004–2005	300	2.4	0.63	Cai et al. (2013)
	2005–2006	300	3.0	0.95	
	2006–2007	300	2.9	0.88	
	2008–2009	300	1.4	0.39	Ding et al. (2013)

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 N_2O emissions from agricultural fields could be expressed in a more general form:

$$F_{\text{annual}} = AX_1^n + F/BX_2 + C, \tag{7}$$

where n, A, B and C are empirical constants, which can be estimated with field measurements, and F is the annual application rate of N-fertilizer. The empirical equation in this study was similar to that established by Lu et al. (2006), who investigated the N_2O emissions from upland soil between 1982 and 2003 in the literature, and also deduced a model including background and fertilizer-induced N_2O emissions based on annual precipitation and fertilizer N input. Nevertheless, unlike our result, Lu et al. (2006) reported a linear regression expression between participation and background N_2O emission. To verify the applicability of the above algorithm, more field studies in various agricultural fields are needed. If the applicability of the above algorithm were widely verified, the regional, or even global, annual cumulative N_2O emissions from agricultural fields could be easily estimated just based on fertilization rates and rainfall in different regions.

4.2 Comparison with previous studies

The results of studies about maize–wheat rotation field in the NCP are shown in Table 4. It is evident that there are very large temporal–spatial variations of the cumulative N_2O emissions and EF_d reported in the NCP. With only the exception of the data in the 2009–2011 maize–wheat rotations, the cumulative N_2O emissions from the NP treatment in this study were 33–108 % greater than the upper limit value reported in the literature. The EF_d value of 0.60 % in 2009–2010 was in good agreement with the values reported by Dong et al. (2000), Ding et al. (2007), Cui et al. (2012), Cai et al. (2013) and Yan et al. (2013), and of 1.1 % in 2010–2011 was in line with the values reported by Li et al. (2010), Wang et al. (2008), Cai et al. (2013) and Yan et al. (2013).

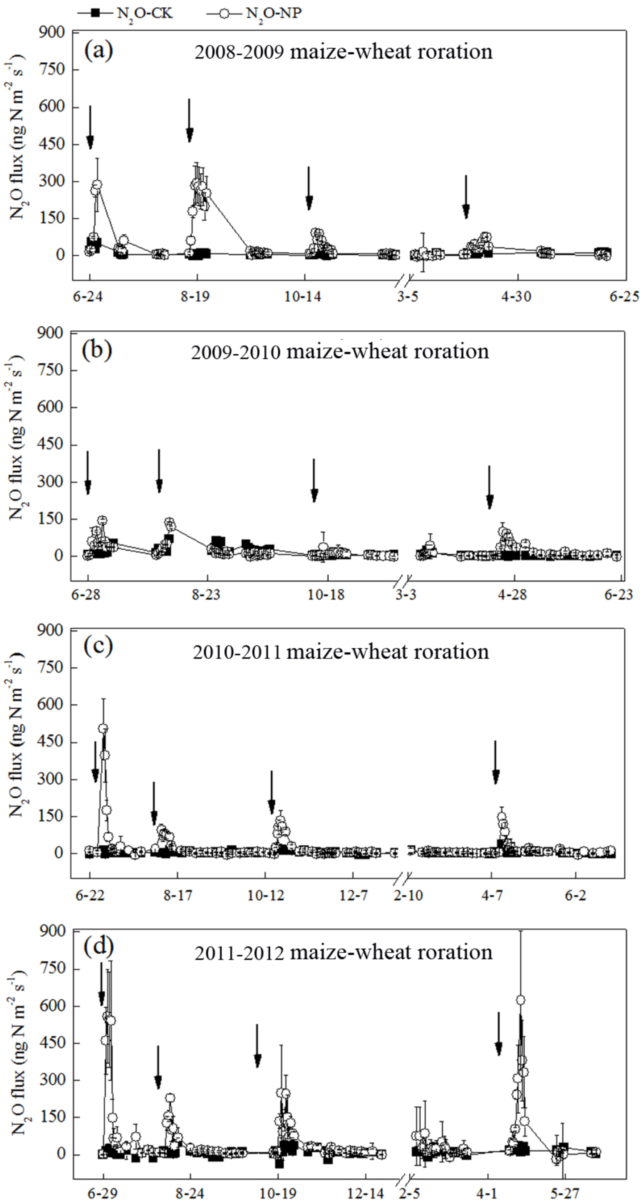

Fig. 2. N_2O emissions from the CK and NP treatments during the N_2O measurement periods in the 2008–2009 **(a)**, 2009–2010 **(b)**, 2010–2011 **(c)** and 2011–2012 **(d)** maize–wheat rotations. Arrows show fertilizer applications.

The EF_d's from the NP treatment 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). In comparison with 2009–2010 and 2010–2011, as shown in Figs. 1 and 2, the higher N_2O emissions in 2008–2009 and 2011–2012 were mainly ascribed to the rain events with relatively high frequency or great intensity just around fertilization events. In addition, the relatively high sampling frequency conducted in this study may be partially responsible for the higher EF_d's. Smith and Dobbie (2001) investigated the impact of sampling frequency on cumulative N_2O fluxes by manual chambers with sampling intervals of 3–7 days and auto-chambers with sampling intervals of 8 hours, and found that the short-lived N_2O peaks after fertilization can not be detected by manual sampling under low sampling frequency. The sampling frequency in this study was each day with a duration at least 10 days after each fertilization event, whereas the sampling frequencies for most previous studies in the NCP were 1–2 times weekly. On the other hand, the very good linear ($R^2 = 0.9996$) response to N_2O concentration (0.093–1.97 ppm) of the GC-ECD improved by our group could make sure the accurate quantification of N_2O in the air samples with remarkably different N_2O concentrations. Most of commercial instruments of GC-ECD have been found to be a non-linear response to N_2O concentrations (Hall et al., 2007; Zheng et al., 2008; Fang et al., 2010; Wang et al., 2010), and thus the single-point calibration for N_2O flux measurement predominately used by previous studies would probably result in relatively low EF_d's.

To check the possible influence of the soils from different areas in the NCP on N_2O emission, soil samples were collected from four sampling sites (Fengqiu, Luancheng, Yucheng and Beijing) where N_2O emissions have been investigated. N_2O emissions from the four fields were simultaneously measured under the same fertilization and irrigation management at the agricultural field of this study (data not shown), and no remarkable difference of N_2O emissions from the four agricultural soils was found in comparison with the uncertainty of the triplet replicates for the agricultural soil investigated in this study. Therefore, the very large temporal–spatial variations of the cumulative N_2O emissions and EF_d's from the agricultural fields in the NCP might also be partially ascribed to the different weather conditions (especially rainfall as mentioned above) in different areas and years during the investigations. To some extent, the field simulation experiment confirmed that the results investigated at any agricultural fields in the NCP could be applied for estimating the annual cumulative N_2O emission and the fertilizer-induced N_2O emission from the agricultural field in the NCP. The estimation would be more representative based on the average value of many years' investigations, because the multi-year rainfall in one small region might partially reflect the uneven distribution of rainfall in different areas of the NCP.

5 Conclusions

Large interannual variations of N_2O emissions were observed from the maize–wheat field in the NCP during the 4 successive years. Precipitation was primarily responsible for the interannual variation of N_2O emission. The significant correlation between cumulative N_2O emission and precipitation obtained in this study may provide an approach to estimate N_2O emission from the area where the rainfall and fertilization rate are similar to this study.

Acknowledgements. This study was funded by the National Science and Technology Pillar Program (no. 2013BAD11B03), the Strategic Priority Research Program of the Chinese Academy of Sciences (no. XDB05010100), the Chinese National Natural Science Foundation (no. 41075094 and 21177140), and the National Basic Research and the Development Program 973 (no. 2010CB732304).

Edited by: X. Wang

References

Barton, L., Kiese, R., Gatter, D., Butterbach-Bahl, K., Buck, R., Hinz, C., and Murphy, D. V.: Nitrous oxide emissions from a cropped soil in a semi-arid climate, Glob. Change Biol., 14, 177–192, 2008.

Bolle, H. J., Seiler, W., and Bolin, B.: Other greenhouse gases and aerosols, in: The Greenhouse Effect, Climate Change and Ecosystems, SCOPE 29 edited by: Bolin, B., Döös, B. R., Jäger, J., and Warrick, R. A., Wiley, New York, 157–203, 1986.

Bouwman, A. F.: Direct emission of nitrous oxide from agricultural soils, Nutr. Cycl. Agroecosys., 46, 53–70, 1996.

Bouwman, A. F., Boumans, L. J. M., and Batjes, N. H.: Emissions of N_2O and NO from fertilized fields: summary of available measurement data, Global Biogeochem. Cy., 16, 1–13, 2002.

Cai, Y. J., Ding, W. X., and Luo, J. F.: Nitrous oxide emissions from Chinese maize-wheat rotation systems: a 3-year field measurement, Atmos. Environ., 65, 112–122, 2013.

Chapuis-Lardy, L., Wrage, N., Metay, A., Chotte, J. L., and Bernoux, M.: Soils, a sink for N_2O, A review, Glob. Change Biol., 13, 1–17, 2007.

Chen, S., Huang, Y., and Zou, J.: Relationship between nitrous oxide emission and winter wheat production, Biol. Fertil. Soils, 44, 985–989, 2008.

Crutzen, P. J.: The influence of nitrogen oxides on the atmospheric ozone content, Q. J. Roy. Meteorl. Soc., 96, 320–325, , 1970.

Cui, F., Yan, G. X., Zhou, Z. X., Zheng, X. H., and Deng, J.: Annual emissions of nitrous oxide and nitric oxide from a wheat-maize cropping system on a silt loam calcareous soil in the North China Plain, Soil Biol. Biochem., 48, 10–19, 2012.

De Klein, C., Novoa, R. S. A., Ogle, S., Smith, K. A., Rochette, P., Wirth, T. C., McConkey, B. G., Mosier, A., and Rypdal, K.: N_2O Emissions from managed soils, and CO_2 emissions from Lime and Urea Application, in: 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Vol 4—Agriculture, Forestry and Other Land Use, edited by: Eggleston, S., Buendia, L., Miwa, K., Ngara, T., and Tanabe, K., Institute for Global Environmental Strategies, Japan, 11.1–11.54, 2006.

Ding, W., Luo, J., Li, J., Yu, H., Fan, J., and Liu, D.: Effect of long-term compost and inorganic fertilizer application on background N_2O and fertilizer-induced N_2O emissions from an intensively cultivated soil, Sci. Total Environ., 465, 115–124, 2013.

Ding, W. X., Cai, Y., Cai, Z. C., Yagi, K., and Zheng, X. H.: Nitrous oxide emissions from an intensively cultivated maize–wheat rotation soil in the North China Plain, Sci. Total Environ., 373, 501–511, 2007.

Dobbie, K. E. and Smith, K. A.: The effects of temperature, water-filled pore space and land use on N_2O emissions from an imperfectly drained gleysol, Eur. J. Soil Sci., 52, 667–673, 2001.

Dong, Y. S., Dieter, S., Manfred, D., Qi, Y. C., and Zhang, S.: N_2O emissions from agricultural soils in the North China Plain: the effect of chemical nitrogen fertilizer and organic manure, J. Environ. Sci., 12, 463–468, 2000.

Fang, S. X., Zhou, L. X., Zhang, F., Yao, B., Zhang, X. C., Zang, K. P., Xu, L., Liu, L. X., Wen, M., and Gu, S.: Dual channel GC system for measuring background atmospheric CH_4, CO, N_2O and SF_6, Acta Scientiae Circumstantiae, 30, 52–59, 2010 (in Chinese).

Firestone, M. K. and Davidson, E. A.: Microbiological basis of NO and N_2O production and consumption in soil, in: Exchange of Trace Gases between Terrestrial Ecosystems and the Atmosphere, edited by: Andreae, M. O. and Schimel, D. S., Wiley, Chichester, 7–21, 1989.

Hall, B. D., Dutton, G. S., and Elkins, J. W.: The NOAA nitrous oxide standard scale for atmospheric observations, J. Geophys. Res., 112, D09305, doi:200710.1029/2006JD007954, 2007.

Halvorson, A. D., Del Grosso, S. J., and Reule, C. A.: Nitrogen, tillage, and crop rotation effects on nitrous oxide emissions from irrigated cropping systems, J. Environ. Qual., 37, 1337–1344, 2008.

Hao, X., Chang, C., Carefoot, J. M., Janzen, H. H., and Ellert, B. H.: Nitrous oxide emissions from an irrigated soil as affected by fertilizer and straw management, Nutr. Cycl. Agroecosys., 60, 1–8, 2001.

Henault, C., Devis, X., Page, S., Justes, E., Reau, R., and Germon, J. C.: Nitrous oxide emissions under different soil and land management conditions, Biol. Fert. Soils, 26, 199–207, 1998.

Hu, X. K., Su, F., Ju, X. T., Gao, B., Oenema, O., Christie, P., Huang, B. X., Jiang, R. F., and Zhang, F. S.: Greenhouse gas emissions from a wheat–maize double cropping system with different nitrogen fertilization regimes, Environ. Pollut., 176, 198–207, 2013.

Huang, Y., Zou, J., Zheng, X., Wang, Y., and Xu, X.: Nitrous oxide emissions as influenced by amendment of plant residues with different C:N ratios, Soil Biol. Biochem., 36, 973–981, 2004.

Khalil, M. I., Schmidhalter, U., and Gutser, R.: N_2O, NH_3 and NO_X emissions as a function of urea granule size and soil type under aerobic conditions, Water Air Soil Poll., 175, 127–148, 2006.

Li, H., Qiu, J., Wang, L., Tang, H., Li, C., and Van Ranst, E.: Modelling impacts of alternative farming management practices on greenhouse gas emissions from a winter wheat-maize rotation system in China, Agr. Ecosyst. Environ., 135, 24–33, 2010.

Liu, C. M., Yu, J. J., and Kendy, E.: Groundwater exploitation and its impact on the environment in the North China Plain, Water Int., 26, 265–272, 2001.

Liu, X. H. and Mu, Z. G. (Eds.): Cropping Systems in China, Agricultural Press of China, Beijing, 1993 (in Chinese).

Lu, Y., Huang, Y., Zou, J., and Zheng, X.: An inventory of N_2O emissions from agriculture in China using precipitation-rectified emission factor and background emission, Chemosphere, 65, 1915–1924, 2006.

Mahmood, T., Ali, R., Malik, K. A., and Shamsi, S. R. A.: Nitrous oxide emissions from an irrigated sandy-clay loam cropped to maize and wheat, Biol. Fertil. Soils, 27, 189–196, 1998.

Meixner, F. X. and Yang, W. X.: Biogenic emissions of nitric oxide and nitrous oxide from arid and semi-arid land, in: Dryland Ecohydrology, edited by D'Odorico, P. and Porporato, A., Springer, Dordrecht, 233–255, 2006.

Meng, L., Ding, W. X., and Cai, Z. C.: Long-term application of organic manure and nitrogen fertilizer on N_2O emissions, soil quality and crop production in a sandy loam soil, Soil Biol. Biochem., 37, 2037–2045, 2005.

Raich, J. W. and Tufekcioglu, A.: Vegetation and soil respiration: correlations and controls, Biogeochemistry, 48, 71–90, 2000.

Rowlings, D. W., Grace, P. R., Kiese, R., and Weier, K. L.: Environmental factors controlling temporal and spatial variability in the soil-atmosphere exchange of CO_2, CH_4 and N_2O from an Australian subtropical rainforest, Glob. Change Biol., 18, 726–738, 2012.

Ruser, R., Flessa, H., Russow, R., Schmidt, C., Buegger, F., and Munch, J. C.: Emission of N_2O, N_2, and CO_2 from soil fertilized with nitrate: effect of compaction, soil moisture and rewetting, Soil Biol. Biochem., 38, 263–274, 2006.

Scheer, C., Wassmann, R., Klenzler, K., Lbragimov, N., and Eschanov, R.: Nitrous oxide emissions from fertilized irrigated cotton (*Gossypium hirsutum* L.) in the Aral Sea Basin, Uzbekistan: influence of nitrogen applications and irrigation practices, Soil Biol. Biochem., 40, 290–301, 2008.

Shi, Y., Wu, W., Meng, F., Zhang, Z., Zheng, L., and Wang, D.: Integrated management practices significantly affect N_2O emissions and wheat–maize production at field scale in the North China Plain, Nutr. Cycl. Agroecosystems, 95, 203–218, 2013.

Smith, K. A.: The potential for feedback effects induced by global warming on emissions of nitrous oxide by soils, Glob. Change Biol., 3, 327–338, 1997.

Smith, K. A. and Dobbie, K. E.: The impact of sampling frequency and sampling times on chamber-based measurements of N_2O emissions from fertilized soils, Glob. Change Biol., 7, 933–945, 2001.

Smith, K. A., Ball, T., Conen, F., Dobbie, K. E., Massheder, J., and Rey, A.: Exchange of greenhouse gases between soil and atmosphere: interactions of soil physical factors and biological processes, Eur. J. Soil Sci., 54, 779–791, 2003.

Sun, Y. L., Lu, P. L., Li, J., Yu, Q., Sun, S. B., Wang, J. S., and Ouyang, Z.: Characteristics of soil N_2O flux in a winter wheat-summer maize rotation system in North China Plain and analysis of influencing factors, Chinese J. Agrometeorol., 29, 1–5, 2008 (in Chinese).

Wang, L. G., Li, H., and Qiu, J. J.: Characterization of Emissions of Nitrous Oxide from Soils of Typical Crop Fields in Huang-Huai-Hai Plain, Scientia Agricultura Sinica, 41, 1248–1254, 2008 (in Chinese).

Wang, Y., Xue, M., Zheng, X., Ji, B., Du, R., and Wang, Y.: Effects of environmental factors on N_2O emission from and CH_4 uptake by the typical grasslands in the Inner Mongolia, Chemosphere, 58, 205–215, 2005.

Wang, Y., Wang, Y., and Ling, H.: A new carrier gas type for accurate measurement of N_2O by GC-ECD, Adv. Atmospheric Sci., 27, 1322–1330, 2010.

Wang, Y. Y., Hu, C. S., Cheng, Y. S., Zhang, Y. M., Ming, H., and Yang, P. P.: Carbon sequestrations and gas regulations in summer-maize and winter-wheat rotation ecosystem affected by nitrogen fertilization in the piedmont plain of Taihang Mountains, China, J. Agro-Environ. Sci., 28, 1508–1515, 2009 (in Chinese).

Yamulki, S., Goulding, K. W. T., Webster, C. P., and Harrison, R. M.: Studies on NO and N_2O fluxes from a wheat field, Atmos. Environ., 29, 1627–1635, 1995.

Yan, G., Zheng, X., Cui, F., Yao, Z., Zhou, Z., Deng, J., and Xu, Y.: Two-year simultaneous records of N_2O and NO fluxes from a farmed cropland in the northern China plain with a reduced nitrogen addition rate by one-third, Agric. Ecosyst. Environ., 178, 39–50, 2013.

Zeng, J. H., Wang, Z. P., Zhang, Y. M., Song, W. Z., Wang, S. B., and Su, W. H.: Flux of N_2O emission from the fields in a wheat and maize rotation system, Chinese J. Environ. Sci., 16, 32–35, 1995 (in Chinese).

Zhang, Y. M., Chen, D. L., Zhang, J. B., Edis, R., Hu, C. S., and Zhu, A. N.: Ammonia volatilization and denitrification losses from an irrigated maize-wheat rotation field in the North China Plain, Pedosphere, 14, 533–540, 2004.

Zhang, Y. Y., Liu, J. F., Mu, Y. J., Pei, S. W., Lun, X. X., and Chai, F. H.: Emissions of nitrous oxide, nitrogen oxides and ammonia from a maize field in the North China Plain, Atmos. Environ., 45, 2956–2961, 2011.

Zhang, Y. Y., Liu, J. F., Mu, Y. J., Xu, Z., Pei, S. W., Lun, X. X., and Zhang, Y.: Nitrous oxide emissions from a maize field during two consecutive growing seasons in the North China Plain, J. Environ. Sci., 24, 1–9, 2012.

Zhang, Y. Y., Mu, Y. J., Fang, S. X., and Liu, J. F.: An improved GC-ECD method for measuring atmospheric N_2O, J. Environ. Sci., 25, 547–553, 2013.

Zheng, X., Mei, B., Wang, Y., Xie, B., Wang, Y., Dong, H., Xu, H., Chen, G., Cai, Z., Yue, J., Gu, J., Su, F., Zou, J., and Zhu, J.: Quantification of N_2O fluxes from soil–plant systems may be biased by the applied gas chromatograph methodology, Plant Soil, 311, 211–234, 2008.