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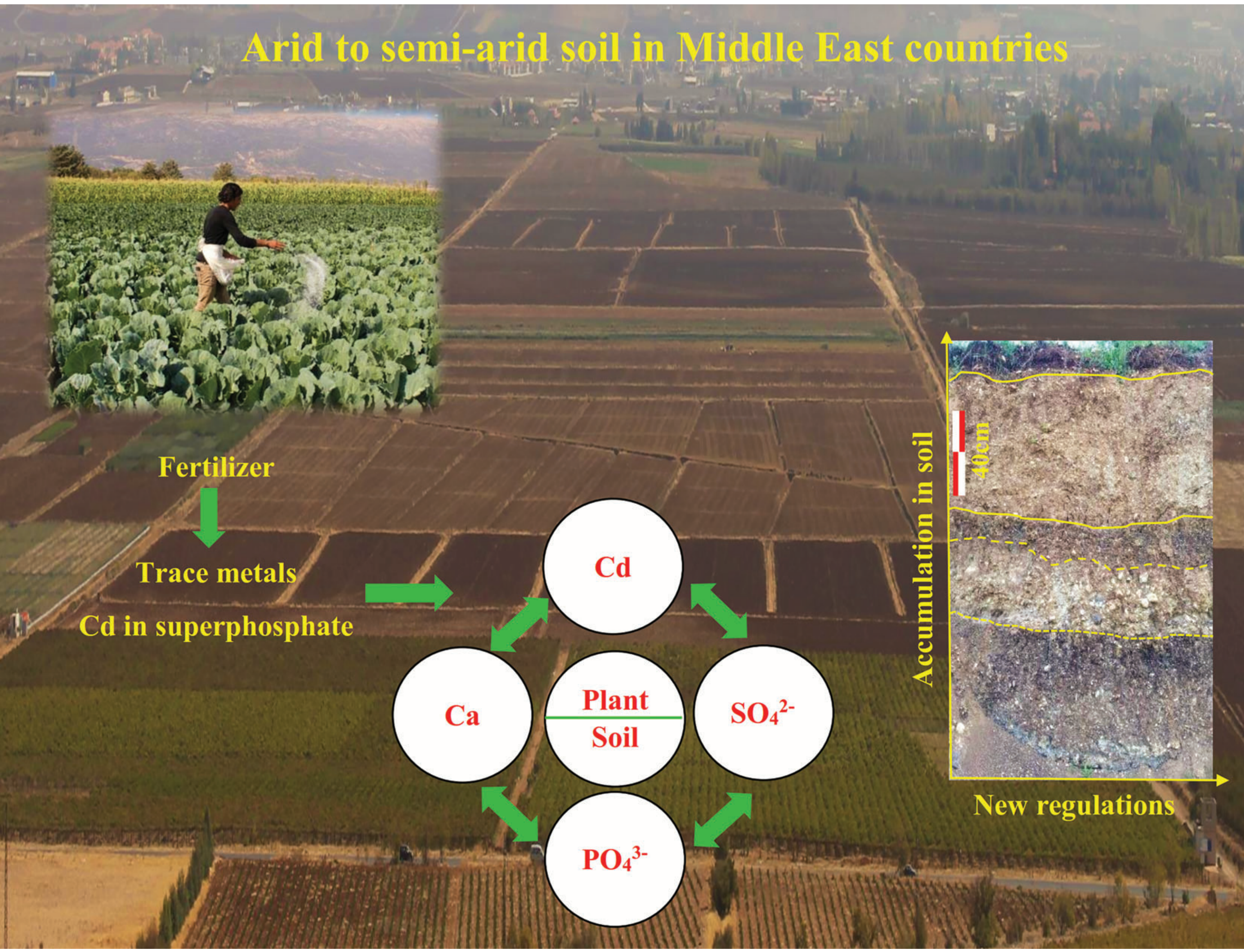

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Research Article

Nitrous Oxide and Methane Emissions in Spring Maize Field in the Semi-Arid Regions of Loess Plateau

A 2-year field study was conducted to measure nitrous oxide (N_2O) and methane (CH_4) in a rain-fed spring maize cropland in the Loess Plateau, P. R. China, and to determine the effects of optimized nitrogen (N) fertilization practices on urea-derived N_2O emission factor (EF), grain yield, net greenhouse gas (NGHG) emission, and net greenhouse gas intensity (NGHGI). Five treatments were considered, including control (CK), conventional N fertilization (Con), optimal N fertilization (Opt), optimal N fertilization plus nitrification inhibitor (Opt + DCD), and optimal N fertilization with slow release urea (Opt + SR). Soil acted as a small sink for atmospheric CH_4. Nitrogen fertilization and heavy rainfall events (>40 mm) were the main factors controlling N_2O emissions. The annual mean EF ranged from 0.12 to 0.55%. Compared to conventional N fertilizer, nitrification inhibitor decreased the annual cumulative N_2O, NGHG, and NGHGI emissions by 45, 52, and 48%, respectively, without decreasing grain yield. In conclusion, nitrification inhibitor addition was the most effective practice to reduce N_2O emissions in the rain-fed regions of Loess Plateau.

Keywords: Nitrification inhibition; Nitrogen practice; Nitrous oxide; Slow release urea; Spring maize

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1 Introduction

Agriculture constitutes an important source of nitrous oxide (N_2O) and methane (CH_4) [1], accounting for approximately 60 and 50% of global N_2O and CH_4 emissions, respectively [2]. Global annual N_2O emissions from nitrogen (N) fertilizers and manures, crop residues, and other agricultural sources are estimated to be up to 4.1 Tg $N_2O{-}N\,y^{-1}$ (range: 3.8–6.8) [3]. Thus, there is an urgent need to reduce N_2O and CH_4 emissions from croplands to mitigate global greenhouse gas (GHG) emissions.

N_2O emissions from cropland increase with an increase in N fertilizer rate [4–6]. Many N fertilization practices, such as balanced N fertilization [7, 8], nitrification inhibitor (NI) [8, 9], and slow release (SR) urea [10, 11], have been proposed to reduce GHG emissions from N-fertilized croplands. However, it is important to note that the effectiveness of these N fertilization practices in reducing N_2O emissions differs greatly, depending on the environmental conditions and farming systems. For instance, some studies showed that NI reduced N_2O emissions by 50–82% in comparison with conventional urea [12, 13]; whereas other studies showed that NI had a limited impact on N_2O emissions [14, 15]. Similarly, SR was found to be able to significantly reduce N_2O emissions from grassland in UK [16], but increase N_2O emissions in the wheat and maize croplands in eastern China [11]. These conflicting results indicate that the mechanism underlying the effects of NI and SR on N_2O emissions remains poorly understood. In addition, upland soils act as a weak sink for atmospheric CH_4 [17], but N fertilization has a variable effect on CH_4 emissions [18]. Some studies reported that N application inhibited atmospheric CH_4 uptake by soils [8, 17], whereas other studies concluded that fertilizer N rates had no significant effect on CH_4 uptake [19]. Thus, more field experiments are required to better understand the effect of different N fertilization practices on N_2O and CH_4 emissions.

GHG emissions are strongly controlled not only by temperature, but also by soil moisture and soil mineral N [20–23]. The content and transformation of soil mineral N could directly influence

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Abbreviations: **ANOVA**, one-way analysis of variance; **CK**, control; **Con**, conventional; **CV**, coefficient of variation; **DCD**, dicyandiamide; **ECD**, electron capture detector; **EF**, emission factor; **FID**, flame ionization detector; **GHG**, greenhouse gas; **IPCC**, Intergovernmental Panel on Climate Change; **LSD**, least significant difference; **NGHG**, net greenhouse gas emission; **NGHGI**, NGHG intensity; **NI**, nitrification inhibitor; **Opt**, optimal; **SR**, slow release; **TN**, total nitrogen; **WFPS**, soil water-filled pore space.

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nitrification and denitrification [5]. In the arid and semi-arid regions, rainfall caused the fluctuation of soil moisture, thus, affecting the microbial activity related to N_2O and CH_4 emissions and the transformation of soil mineral N, and consequently the N_2O and CH_4 emissions [11, 21]. The emission factor (EF) was used to estimate N_2O emissions on the basis of the amount of N fertilizer used in a country or geographic region, usually using the default factor (1 or 1.25% derived from Intergovernmental Panel on Climate Change (IPCC) [24] and a meta-analysis of EF in global cropland [25], respectively). However, it is noted that arid and semi-arid regions are not included in the above meta-analysis. In addition, previous empirical studies have shown that EF is not constant, but varies from 0.1 to 73.7% in different crops, countries, and regions [26]. Thus, an accurate EF is needed for accurate estimation of N_2O emissions in arid and semi-arid regions.

In the Loess Plateau, P. R. China, the soil fertility in general and the soil N level in particular are very low, with a total nitrogen (TN) content of 0.06–0.08% [27, 28]. Therefore, winter wheat and spring maize, the two main crops in this region, are heavily fertilized with chemical N fertilizer (200 and 230 kg N ha^{-1}, respectively) during the past decades to obtain a high yield [29–31]. Rainfall is the sole source of soil water supply. In this region, the annual precipitation for the period 1995–2004 ranged from a minimum of 332 mm in 1995 to a maximum of 919 mm in 2004 with a co-efficient of variation (CV) of 22%, and the precipitation from July to September ranged from 147 to 609 mm with a CV of 37% [32]. As the effect of N fertilization depends critically on water availability in the arid and semi-arid regions [33, 34], N fertilizer tends to be of low efficiency in the Loess Plateau, resulting in potential environmental risks such as GHG emissions [35, 36] and subsoil nitrate accumulation and leaching [37, 38]. However, there have been few studies investigating EF and the main factors controlling N_2O and CH_4 emissions in rain-fed farming regions.

N_2O and CH_4 emissions from a rain-fed spring maize cropland in the semi-arid Loess Plateau were measured in this 2-year field study. The objectives were to (i) quantify N_2O and CH_4 emissions after various N fertilization practices; (ii) identify EFs and the main factors controlling N_2O and CH_4 emissions; and (iii) identify the most effective N fertilization practice in the semi-arid Loess Plateau, P. R. China.

2 Materials and methods

2.1 Site description

A field experiment was carried out at State Key Agro-Ecological Experimental Station in the Loess Plateau (35°12′N, 107°40′E; 1220 m asl) in Changwu County, Shaanxi Province, P. R. China. The study area is a typical rain-fed farming region and has a semi-arid continental monsoon climate. The mean annual rainfall from 1984 to 2014 was 560 mm, about 60% of which falls between June and September. The open pan evaporation is 1440 mm. The mean annual temperature is 9.4°C. The daily air temperature and precipitation during the study period from January 2013 to December 2014 are shown in Fig. 1.

The soil at the study site is loam (Cumulic Haplustoll; USDA Soil Taxonomy System) developed from loess deposits, and contains 8% sand, 70% silt, and 22% clay [39]. Soils in the top 20 cm are composed of $CaCO_3$ 10.5%, organic C 6.5 g kg^{-1}, and TN 0.80 g kg^{-1}, with a field water holding capacity of 0.29 cm^3 cm^{-3}, a pH of 8.4 (soil: H_2O suspension, 1:1), and a bulk density of 1.3 Mg m^{-3}.

2.2 Experimental design and crop management

A high-yield spring maize (*Zea mays* L.) hybrid (Pioneer, 335), a representative crop type in this region, was chosen for this field study. There were five treatments with three replicates each in a completely randomized design, giving a total of 15 plots. All plots were 5.5 × 18 m with a spacing of 0.5 m between adjacent plots and 1 m between adjacent blocks. The treatments were as follows:

(1) Control (CK): No fertilizer N applied.
(2) Conventional N fertilizer (Con): Urea (N 46%) applied at a rate of 200 kg N ha^{-1} y^{-1}, which is the general practice in this region.
(3) Optimum N fertilizer (Opt): Urea applied at a rate of 160 kg N ha^{-1} y^{-1} based on the recommendation of local extension service.
(4) Optimum N fertilizer plus NI (Opt + DCD): Urea and solid dicyandiamide (DCD, chemically pure, Sinopharm Chemical Reagents) applied at a rate of 10% N, and broadcast onto the soil surface before plough.
(5) Optimum N fertilizer with slow release urea (Opt + SR): SR is coated urea (urea formaldehyde) containing 26% N.

Phosphorus (superphosphate) and potassium (potassium sulfate) fertilizers were applied at a rate of 117 kg P_2O_5 ha^{-1} and 37.5 kg K_2O ha^{-1} before seeding. All fertilizers were mixed into the top 20 cm soil as basal fertilizer by ploughing. Soil was covered with plastic film (750 mm wide by 0.008 mm thick) in order to reduce soil evaporation and increase soil temperature, and crop growth. The plant density was 57 000 plants ha^{-1} at a distance of 30 cm in rows and 60 cm between rows.

Maize straw was removed from the plots after harvest, and weeds were removed manually during the growing season. No irrigation was applied during the growing season, and thus, soil water supply was completely provided by natural rainfall. Fallow (from September to May of the next year) and maize (from May to September) were rotated at the study site. Maize was planted on April 24 and harvested on September 9 in 2013, and planted on April 30 and harvested on September 15 in 2014.

2.3 Gas sampling and measurements

N_2O and CH_4 emissions were measured using the static chamber method [40]. Two types of static chambers were used in this study, each consisting of base frame and a removable upper chamber made of stainless steel. The base frame and chamber were 60 × 50 × 20 cm (length × width × height) and 60 × 50 × 50 cm for type I chamber; and 60 × 30 × 20 cm and 60 × 30 × 20 cm (length × width × height) for type II chamber, respectively. Prior to planting, the base frames were inserted 20 cm into the soil (half mulching soil and half bare soil), and then remained in place throughout the measurement period except for tillage, which was performed once a year. A visual description is shown in Fig. 2. Type I chamber was used to measure N_2O emissions when maize height was <50 cm, with two maize plants in each chamber; while type II chamber was used when maize height was >50 cm. Type II chamber was separated vertically into two parts with a hole (11 cm in diameter) at the top center of the chamber, and only one maize plant was placed in the chamber [41].

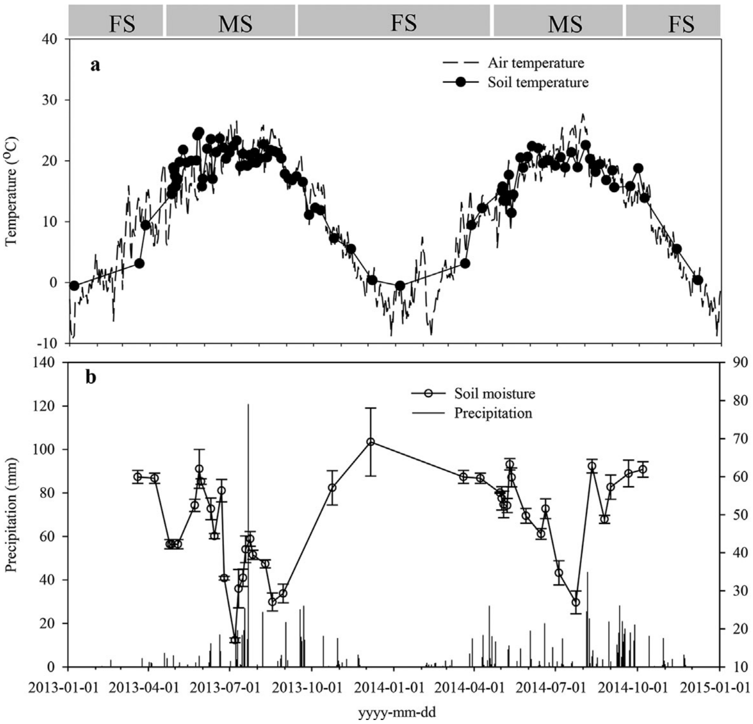

Figure 1. Air and soil temperature at 0–20 cm depths (a) and precipitation and soil WFPS at 0–20 cm depths (b). (FS, fallow season; MS, maize growing season).

This allowed the cornstalks to pass through the chamber and as a result, to only cover the maize root. The gap between the chamber and the cornstalk was sealed using a 1.2 μm-thick preservative film made of polyvinylidene chloride when the chamber was closed. All chambers were sealed with rubber and covered with an insulating layer to minimize chamber effects on air temperature to <3°C in the headspace during gas sampling, and two opposing ventilators were installed inside the chamber to ensure complete mixing of air.

Gas samples were collected using 50 mL plastic injectors through a three-way stopcock and a Teflon tube connected to the chamber at 0, 10, 20, 30, 40 min after the chambers were closed. Under normal circumstances, N_2O and CH_4 emissions were measured once a week during the growing season of maize, once a month when the soil was frozen (from November to March of next year), and every 10 days for the fallow season. However, daily measurements were carried out for about 10 days after fertilizer application and 3 days after heavy rainfall events (>20 mm), respectively. All measurements were conducted between 08:00 a.m. and 11:00 a.m. local time, and there were a total of 57 and 40 measurements in 2013 and 2014, respectively.

Gas samples were analyzed for N_2O and CH_4 within 24 h of sampling using gas chromatography (Agilent GC6820, Agilent, USA) equipped with an electron capture detector (ECD) and a flame ionization detector (FID). Temperatures in the ECD and column oven were 300 and 60°C, respectively. Pure N_2 (99.999%) was used as the carrier gas and 10% CO_2 in pure N_2 as the buffer gas for ECD, respectively. The detection limit was $2\,\mu g\,N\,m^{-2}\,h^{-1}$ for N_2O and $3.4\,\mu g\,C\,m^{-2}\,h^{-1}$ for CH_4 when the chamber height was 50 cm. The gas samples were calibrated using compressed air ($333\,\mu L\,m^{-3}$ N_2O and $197\,\mu L\,m^{-3}$ CH_4) during each measurement cycle.

N_2O and CH_4 emission rates were calculated from the linear increase in the concentration in the chamber during the sampling period. As all measurements of CH_4 emissions were negative, CH_4 uptakes (positive) were used for convenience in this study. The cumulative N_2O emissions and CH_4 uptakes were estimated using linear interpolation between every two adjacent intervals of the measurements (Eqs. (1) and (2)) [37].

$$N_{cum} = \sum_{i=first}^{i=last-1}\left(\left(\frac{D_i - D_{i+1}}{2}\right) \times (N_{i+1} - N_i - 1) + D_i\right) + D_{last} \tag{1}$$

$$D_i = F \times 24 \times 10^{-5} \tag{2}$$

where N_{cum} ($kg\,ha^{-1}\,y^{-1}$) is the annual N_2O cumulative emissions or CH_4 cumulative uptakes; D_i ($kg\,ha^{-1}$ per day) is the daily N_2O cumulative emissions or CH_4 cumulative uptakes; first and last are the first and last measurements of N_2O emissions or CH_4

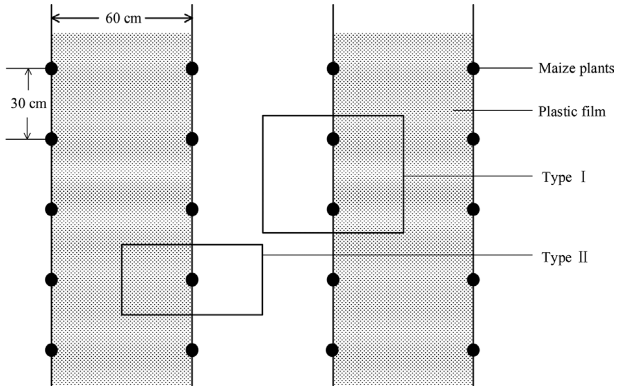

Figure 2. Schematic diagram of the installation of base frames. Black dots denote the maize plants. Shadow denote the plastic film. Types I and II denote the two types of chambers used in this study.

uptakes; $N_{i+1}-N_i-1$ is the interval between two adjacent measurements; F ($\mu g\ m^{-2}\ h^{-1}$) is the N_2O emission rate or CH_4 uptake rate; 24 and 10^{-5} are the conversion co-efficients.

2.4 Soil temperature, soil moisture, NO_3-N, and NH_4-N

Air temperature inside the chamber and soil temperature at a 10 cm depth were measured using a portable digital thermometer (JM624, Tianjin Jinming Instruments, P. R. China) immediately after the first and last sampling, and the mean of the two measurements was used as the temperature on the sampling day. Soil samples at a 20 cm depth were collected once every 2 days for 10 days following N fertilization and within 4 days following heavy rainfalls. Over the remaining period, soils were sampled once when gases were sampled. Gravimetric soil water content w/w was measured after drying the soil in an oven at 105°C for 24 h, and soil water-filled pore space (WFPS) was calculated using Eq. (3). To determine NH_4-N and NO_3-N content, fresh samples (24 g) were extracted using 100 mL of 1 mol L^{-1} KCl solution, and then the extracts were analyzed using continuous flow analysis (TRAACS2000, Bran and Luebbe, Norderstedt, Germany).

$$\mathrm{WEPS} = \frac{\text{Soil water content (\%)} \times \text{Soil bulk density}}{1 - \frac{\text{Soil bulk density}}{2.65}} \times 100\% \qquad (3)$$

2.5 Grain yield

Maize was manually harvested from an area of 16 m^2 in each plot. The samples were dried at 65°C to a constant weight to determine the aboveground biomass. The grain yield was expressed at 15.5% moisture [42].

2.6 Net greenhouse gas emission (NGHG), net greenhouse gas emission intensity (NGHGI), and EF

NGHG emissions were calculated in terms of N_2O plus CH_4 fluxes in CO_2 equivalents (CO_2-eq) to evaluate the effects of different N fertilization practices on N_2O emissions and CH_4 uptakes. The global warming potentials of 1 kg N_2O and CH_4 being equivalent to 298 and 25 kg CO_2 at the 100-year time horizon, respectively [41], were applied to measure NGHG (kg $CO_2-eq\ ha^{-1}$) in CO_2-eq. NGHGI (kg $CO_2-eq\ Mg^{-1}$ grain) was calculated by dividing NGHG by crop yield and expressed as the magnitude of NGHG to produce the same crop yield. EF was determined using Eq. (4).

$$\mathrm{EF} = \sum_{i=\mathrm{first}}^{i=\mathrm{last}-1}\left(\left(\frac{D_i - D_{i+1}}{2}\right) \times (N_{i+1} - N_i - 1) + D_i\right) + D_{\mathrm{last}}$$

$$\mathrm{EF} = \frac{N_{\mathrm{tr-cum}} - N_{\mathrm{ck-cum}}}{N_{\mathrm{rate}}} \times 100\% \qquad (4)$$

where $N_{\text{tr-cum}}$ (kg $N_2O-N\ ha^{-1}\ y^{-1}$) is the annual N_2O cumulative emissions in the four N treatments, $N_{\text{ck-cum}}$ (kg $N_2O-N\ ha^{-1}\ y^{-1}$) is the annual N_2O cumulative emissions in CK treatment, and N_{rate} (kg N $ha^{-1}\ y^{-1}$) is the N fertilization rate in the four N treatments.

2.7 Data analysis

All data in this study are presented as mean ± SD. In each year, the differences in grain yield, seasonal, and annual N_2O and CH_4 cumulative emissions among the five treatments were analyzed using one-way analysis of variance (ANOVA) followed by least significant difference (LSD) post-hoc test. A p-value of <0.05 was considered significant. Pearson correlation analysis was performed to determine the relationships between N_2O emissions and soil variables. All statistical analyses were performed using SPSS 17.0 for Windows (SPSS Statistics, Chicago, IL, USA).

3 Results

3.1 Grain yield

Nitrogen fertilization significantly enhanced maize grain yield in both 2013 and 2014 ($p < 0.05$; Tab. 1). The maize yield for Opt, Opt + DCD, and Opt + SR treatments ranged from 9.61 to 10.46 Mg ha^{-1} in 2013 with an increase of 31.6–43.3%; and from 11.41 to 11.92 Mg ha^{-1} in 2014 with an increase of 185–198% in comparison to CK treatment, respectively. However, there was no significant difference in the grain yield between N optimized (Opt, Opt + DCD, and Opt + SR) treatments and Con treatment (11.3 Mg ha^{-1}) in both 2013 and 2014, although the N fertilization rate of the former was 20–25% lower than that of the latter.

Table 1. Yield, NGHG, and NGHGI for different N fertilization practices in 2013 and 2014

Treatment	2013			2014		
	Yield (Mg ha^{-1})	NGHG (kg $CO_2-eq\ ha^{-1}$)	NGHGI (kg $CO_2-eq\ Mg^{-1}$ grain)	Yield (Mg ha^{-1})	NGHG (kg $CO_2-eq\ ha^{-1}$)	NGHGI (kg $CO_2-eq\ Mg^{-1}$ grain)
N0	7.30 ± 0.94^{b}	301 ± 15^{d}	42 ± 4^{bc}	3.99 ± 1.41^{b}	240 ± 11^{d}	65 ± 22^{a}
Con	10.30 ± 0.44^{a}	807 ± 53^{a}	78 ± 4^{a}	12.23 ± 0.66^{a}	769 ± 74^{a}	63 ± 9^{a}
Opt	10.46 ± 0.33^{a}	555 ± 72^{b}	53 ± 8^{b}	11.63 ± 2.69^{a}	517 ± 53^{b}	47 ± 18^{ab}
Opt + DCD	9.61 ± 0.84^{a}	383 ± 59^{cd}	40 ± 9^{c}	11.92 ± 0.94^{a}	304 ± 56^{c}	26 ± 6^{b}
Opt + SR	10.10 ± 0.75^{a}	444 ± 51^{c}	44 ± 7^{bc}	11.41 ± 0.54^{a}	438 ± 51^{b}	38 ± 4^{b}

NGHG (kg $CO_2-eq\ ha^{-1}$) = $N_2O-N \times 44/28 \times 298 + CH_4-C \times 16/12 \times 25$.
NGHGI (kg $CO_2-eq\ Mg^{-1}$ grain) = NGHG/yield.
Values are expressed as mean ± standard deviation ($n = 3$). The values within the columns followed by different letters are significantly different at $p < 0.05$.

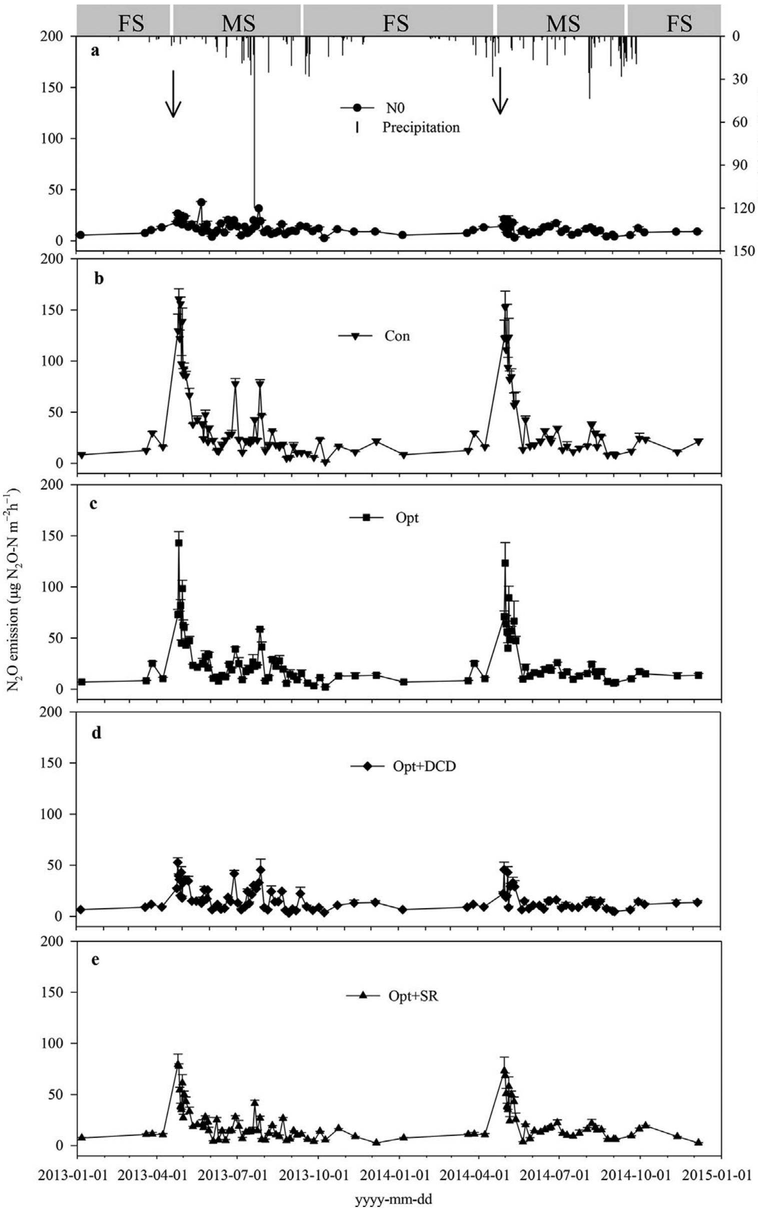

Figure 3. Nitrous oxide emissions (N_2O) from fields with different nitrogen fertilization practices across fallow and maize growing season in 2013 and 2014. (FS, fallow season; MS, maize growing season).

3.2 N_2O emissions

In both years, the N_2O emission rates in all treatments increased rapidly to a peak within 2 days after N fertilization, maintained at a high level for about 10 days, and then decreased rapidly to a lower level of 0–20 $\mu g\,N_2O{-}N\,m^{-2}\,h^{-1}$ (Fig. 3). However, peaks could also be observed after high rainfalls (>40 mm). During the 2-year study period, the mean N_2O emission rate was 28.8, 17.3, and 21.4 $\mu g\,N_2O{-}N\,m^{-2}\,h^{-1}$ for Opt, Opt + DCD, and Opt + SR treatments, with a decrease of 29.5, 57.7, and 47.7% compared with Con treatment (40.9 $\mu g\,N_2O{-}N\,m^{-2}\,h^{-1}$), respectively. In both years, the cumulative N_2O emissions within 10 days after N fertilization accounted for about 13% of the total annual N_2O emissions in CK and Opt + DCD treatments, and about 26% in other treatments, respectively. In 2013, the largest daily N_2O emission was observed on the second day of N fertilization, which was 26.5, 160.7, 143.1, 52.3, and 77.7 $\mu g\,N_2O{-}N\,m^{-2}\,h^{-1}$ in CK, Con, Opt, Opt + DCD, and Opt + SR treatments, respectively. A similar phenomenon was also observed in 2014. N_2O emissions had a strong response to rainfall, especially when the rainfall was >40 mm. For example, N_2O emission peaks were observed after heavy rainfall on July 22, 2013 (120 mm) (31.4, 77.7, 58.5, 33.0, and 26.9 $\mu g\,N_2O{-}N\,m^{-2}\,h^{-1}$ in CK, Con, Opt, Opt + DCD, and Opt + SR, respectively). A similar phenomenon was also observed on August 6 (44 mm) and September 30 (40.6 mm) in 2014. The cumulative N_2O emissions induced by rainfall accounted for 6.4 and 12.5% of the total annual N_2O emissions in 2013 and 2014, respectively.

The annual N_2O emissions in Con, Opt, Opt + DCD, and Opt + SR treatments (0.91–1.88 $kg\,N_2O{-}N\,ha^{-1}\,y^{-1}$) were significantly higher than that of CK treatment (0.80 $kg\,N_2O{-}N\,ha^{-1}\,y^{-1}$) ($p < 0.05$), indicating that N fertilization could significantly increase annual N_2O emissions (Tab. 2). However, the annual N_2O emission in Opt, Opt + DCD, and Opt + SR treatments was decreased by 27.4, 45.2, and 39.6% in 2013, and by 27.7, 51.6, and

Table 2. Cumulative N_2O emission ($kg\ N_2O{-}N\ ha^{-1}$) and CH_4 uptake ($kg\ CH_4{-}C\ ha^{-1}$) for different N fertilization practices in 2013 and 2014

Treatment	2013 MS	2013 FS	2013 MS + FS	2014 MS	2014 FS	2014 MS + FS
N_2O						
N0	0.52 ± 0.01^d	0.35 ± 0.01^b	0.87 ± 0.03^d	0.39 ± 0.02^c	0.33 ± 0.01^b	0.72 ± 0.03^c
Con	1.42 ± 0.12^a	0.54 ± 0.05^a	1.97 ± 0.09^a	1.28 ± 0.15^a	0.60 ± 0.06^a	1.88 ± 0.14^a
Opt	1.01 ± 0.10^b	0.42 ± 0.05^b	1.43 ± 0.14^b	0.90 ± 0.06^b	0.46 ± 0.06^b	1.36 ± 0.12^b
Opt + DCD	0.68 ± 0.06^c	0.40 ± 0.05^b	1.08 ± 0.11^c	0.50 ± 0.07^c	0.41 ± 0.03^b	0.91 ± 0.10^c
Opt + SR	0.79 ± 0.11^c	0.39 ± 0.01^b	1.19 ± 0.11^c	0.77 ± 0.13^b	0.43 ± 0.01^b	1.20 ± 0.13^b
CH_4						
N0	1.88 ± 0.12^a	1.31 ± 0.15^a	3.19 ± 0.26^a	1.65 ± 0.02^a	1.32 ± 0.08^a	2.98 ± 0.09^a
Con	1.84 ± 0.01^a	1.61 ± 0.27^a	3.45 ± 0.26^a	1.57 ± 0.06^a	1.78 ± 0.31^a	3.35 ± 0.28^a
Opt	1.80 ± 0.05^a	1.66 ± 0.14^a	3.46 ± 0.17^a	1.86 ± 0.07^a	1.74 ± 0.27^a	3.60 ± 0.23^a
Opt + DCD	1.93 ± 0.05^a	1.74 ± 0.32^a	3.67 ± 0.28^a	1.91 ± 0.05^a	1.73 ± 0.32^a	3.64 ± 0.37^a
Opt + SR	1.62 ± 0.10^a	1.73 ± 0.05^a	3.35 ± 0.11^a	1.83 ± 0.09^a	1.89 ± 0.13^a	3.72 ± 0.09^a

FS, fallow season; MS, maize growing season.
Values are expressed as mean ± standard deviation ($n = 3$). The values within the columns followed by different letters are significantly different at $p < 0.05$ in N_2O or CH_4.

36.2% in 2014 compared with Con treatment (1.88 $kg\ N_2O{-}N\ ha^{-1}$ in 2013 and 1.97 $kg\ N_2O{-}N\ ha^{-1}$ in 2014), respectively. The cumulative N_2O emission during the fallow season accounted for 27.4–40.2% of the total annual N_2O emissions in 2013 and 31.9–45.8% in 2014.

EF values ranged from 0.12 to 0.55%, and were higher than that of growth season (0.07–0.45%). The lowest value was observed in Opt + DCD treatment, followed by Opt + SR, Opt, and Con treatments (Tab. 3).

3.3 CH_4 uptakes

Soils acted as a small sink for atmospheric CH_4 (Fig. 4). There was no significant difference in daily or annual mean CH_4 uptakes among the five treatments ($p > 0.05$). The daily CH_4 uptake ranged from 0.15 to 127 $\mu g\ CH_4{-}C\ m^{-2}\ h^{-1}$ with an average of 45 $\mu g\ CH_4{-}C\ m^{-2}\ h^{-1}$, and the annual CH_4 uptake ranged from 3.0 to 3.7 $kg\ CH_4{-}C\ ha^{-1}$ with an average of 3.4 $kg\ CH_4{-}C\ ha^{-1}$ (Tab. 2). CH_4 uptakes increased from April to July and then decreased slowly in winter.

3.4 Soil temperature, WFPS, $NO_3{-}N$, and $NH_4{-}N$

Soil temperature at a 10 cm depth (range: −0.5 to 24.7°C; mean: 17.7°C) varied with daily mean air temperature (Fig. 1a). In the top 20 cm soils, soil WFPS markedly increased after heavy rainfall events and then decreased rapidly due to soil texture, high evaporation, and crop uptake (Fig. 1b). It ranged from 17.0 to 70.0% with an average of 48.8%, and was <60.0% for most of the study period.

The $NO_3{-}N$ content showed a high response to N fertilization and heavy rainfalls (>40 mm) (Fig. 5). The average soil $NO_3{-}N$ content was 22, 106, 100, 71, and 61 $mg\ kg^{-1}$ for CK, Con, Opt, Opt + DCD, and Opt + SR treatments, respectively. The $NO_3{-}N$ content increased markedly after N fertilization, and maintained at high level for a relatively long period. Application of NI and SR led to a lower $NO_3{-}N$ content than Opt treatment. Furthermore, the $NO_3{-}N$ content increased rapidly after heavy rainfalls. For example, the $NO_3{-}N$ content in all treatments except CK treatment reached a peak after the heavy rainfall (120 mm) on 22 July, 2013 (217, 276, 37, and 83 $mg\ kg^{-1}$ for Con, Opt, Opt + DCD, and Opt + SR treatments, respectively). The same phenomenon was also observed after a heavy rainfall (44 mm) on August 6, 2014. In contrast, the $NH_4{-}N$ content was relatively low (0.35 to 58.0 $mg\ kg^{-1}$) (Fig. 6). It increased rapidly to a peak within 2 days after fertilization, and then decreased rapidly to a lower level. The mean $NH_4{-}N$ content was 5, 13, 10, 12, and 9 $mg\ kg^{-1}$ for CK, Con, Opt, Opt + DCD, and Opt + SR treatments, respectively. Application of NI and SR led to higher soil $NH_4{-}N$ content than Opt treatment.

3.5 NGHG and NGHGI

Compared with Con treatment, NGHG was decreased by 31.2, 52.5, and 45.0% in Opt, Opt + DCD, and Opt + SR treatments in 2013, and

Table 3. Direct N_2O emission factors (%) for different N fertilization practices in 2013 and 2014

Treatments	2013 MS	2013 MS + FS	2014 MS	2014 MS + FS	Mean MS	Mean MS + FS
Con	0.45 ± 0.05^a	0.55 ± 0.05^a	0.44 ± 0.07^a	0.58 ± 0.07^a	0.45	0.57
Opt	0.31 ± 0.05^b	0.35 ± 0.08^b	0.32 ± 0.03^b	0.40 ± 0.07^b	0.31	0.38
Opt + DCD	0.10 ± 0.04^c	0.13 ± 0.07^c	0.07 ± 0.04^c	0.12 ± 0.06^c	0.08	0.12
Opt + SR	0.17 ± 0.07^c	0.20 ± 0.07^c	0.24 ± 0.07^b	0.30 ± 0.07^b	0.20	0.25

FS, fallow season; MS, maize growing season.
Values are expressed as mean ± standard deviation ($n = 3$). The values within the columns followed by different letters are significantly different at $p < 0.05$.

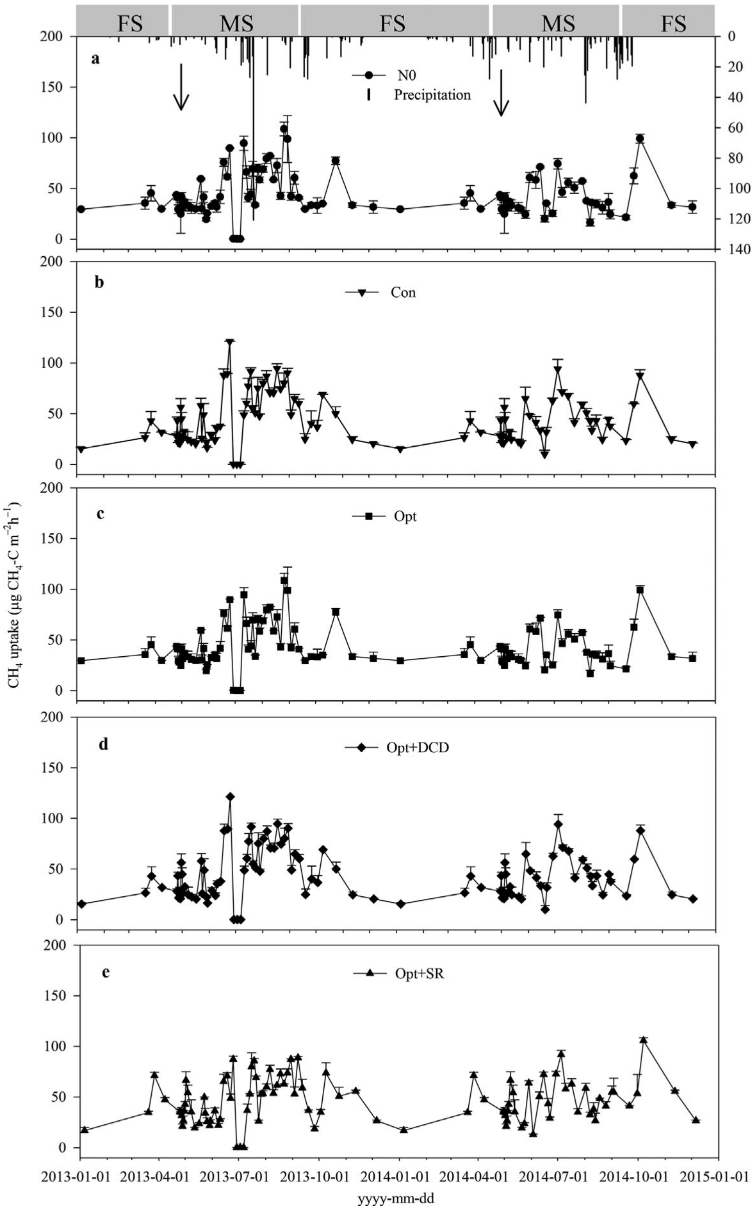

Figure 4. Methane (CH_4) uptake from fields with different nitrogen fertilization practices across fallow and maize growing season in 2013 and 2014. (FS, fallow season; MS, maize growing season).

by 32.8, 60.5, and 43.0% in 2014 ($p < 0.05$) (Tab. 1), respectively; and NGHGI was decreased by 32.1, 48.7, and 43.6% in 2013, and by 25.4, 58.7, and 39.7% in 2014, respectively. The highest decrease occurred in Opt + DCD treatment, followed by Opt + SR treatment.

4 Discussion

4.1 N_2O emissions from croplands in semi-arid regions

The annual cumulative N_2O emissions after N fertilization practices ranged from 0.72 to 1.97 kg N_2O–N ha^{-1} in this study, which fell well within the range reported in a meta-analysis for global croplands (0.3–16.8 kg N_2O–N ha^{-1} y^{-1}) [43] and maize cropland in the Loess Plateau (0.65–3.52 kg N_2O–N ha^{-1} y^{-1}) [44]. However, the mean annual cumulative N_2O emission in this study (1.38 kg N_2O–N ha^{-1} y^{-1}) was significantly lower than that in the irrigated regions [41, 45, 46]. This may be because intensive water management, such as irrigation, was practiced in the irrigated regions, leading to an increase in the frequencies of alternate wetting and drying. Consequently, the C and N mineralization rates increased, which provided enough substrate for N_2O production [22, 37].

The background N_2O emission (0.80 kg N ha^{-1} y^{-1}) fell within the range for various soil/climate regions and major cropping systems in P. R. China (0.1 to 3.67 kg N ha^{-1} y^{-1} with a mean of 1.5 kg N ha^{-1} y^{-1}) [47], and accounted for 38.3–80.0% of the total N_2O emissions in the four N fertilization practices. A higher proportion of 81.8% was reported for a rain-fed wheat cropland in Western Australia [48], but its baseline N_2O emission (0.09 kg N ha^{-1} y^{-1}) was approximately nine times lower than that in this study. The baseline N_2O emission was suggested to be resulting from nitrification and denitrification of soil indigenous N [47, 48]. The TN content (0.8 g kg^{-1}) and precipitation (550 mm) in this

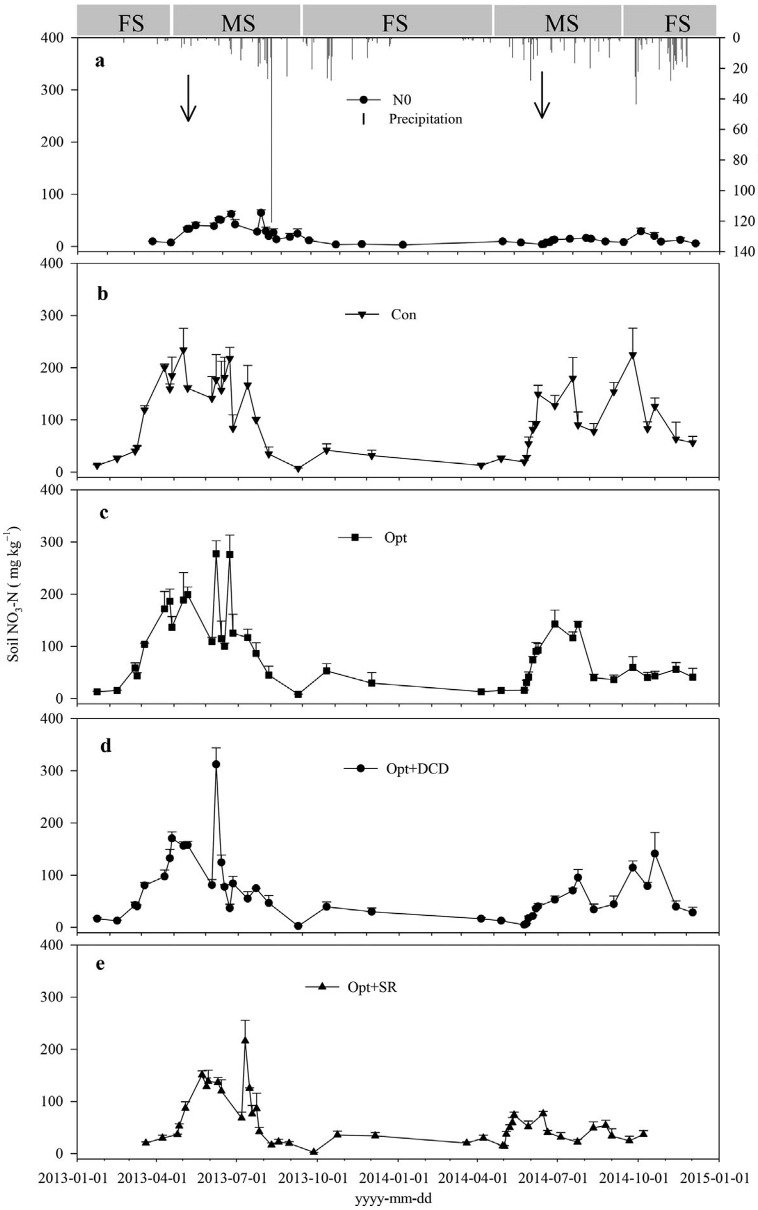

Figure 5. Dynamics of soil nitrate content at 0–20 cm depths from fields with different nitrogen fertilization practices across fallow and maize growing season in 2013 and 2014. (FS, fallow season; MS, maize growing season).

study were higher than that in Western Australia (0.56 g kg^{-1} and 550 mm) [48]. In addition, in this study crop grew in hot and wet summer, but in cold and wet winter in Barton's study [48]. Thus, high TN content, temperature, and precipitation may contribute to the baseline N_2O emissions.

EF was $0.34 \pm 0.18\%$ in this study, which was within the range for fertilized uplands in P. R. China (0.22–1.53%) [49], and the range for maize croplands in Canada (0.03–1.45%) [50]. However, it was much lower than 1% suggested by IPCC [24] and 1.06% suggested by Davidson and Verchot [51]. Barton et al. [48] also reported a low EF (0.02%) in rain-fed croplands in semi-arid Southwestern Australia. Thus, the default value (1%) could cause an overestimation of direct N_2O emissions in the rain-fed cropland in the semi-arid regions, and multi-year measurements at multiple sites were needed to validate the emission factor for rain-fed croplands in semi-arid regions. EF was lower in rain-fed regions than in the irrigated regions, such as North China Plain (0.61–0.77%), probably due to no irrigation and lower precipitation [52].

4.2 Factors controlling N_2O emissions

There was no significant relationship between soil temperature, WFPS, and N_2O emissions in this study, which was in agreement with other studies [8, 19]. However, N_2O emission was closely related to rainfall event (>40 mm) [50, 53], and the peaks of N_2O emissions occurred after heavy rainfall events, such as on July 22, 2013 (120 mm), August 6 (44 mm) 2014, and September 30 (40.6 mm) 2014. After heavy rainfalls, the denitrification rate increased due to the formation of anaerobic environment [54]. In addition, the rates of C and N mineralization, as well as the cell lysis and intercellular solutes [55, 56] increased for several days following the wetting of dry soil [22, 37, 57]. As a result, soil mineral N increased, thus, providing enough substrate for N_2O production. For example, the NO_3–N content after the heavy rainfall on July 20, 2013 was 20–176% higher than that before rainfall. However, no N_2O peaks were observed after the

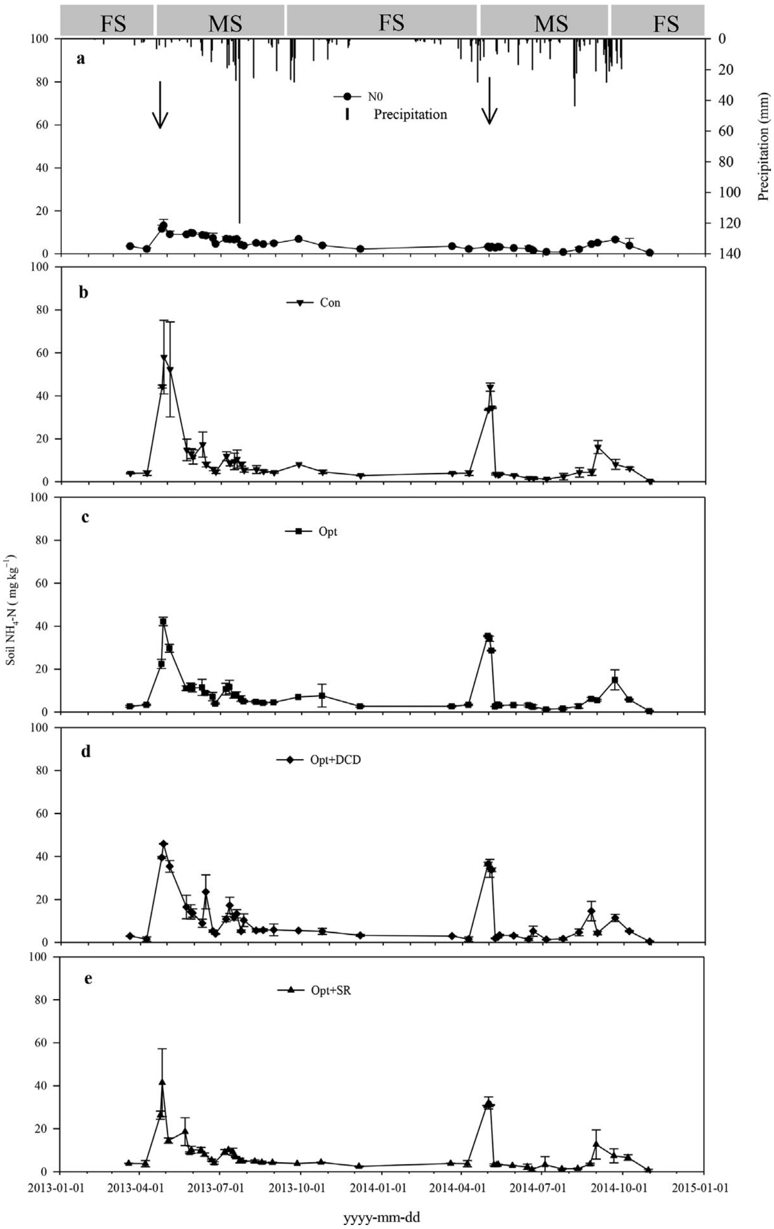

Figure 6. Dynamics of soil ammonium content at 0–20 cm depths from fields with different nitrogen fertilization practices across fallow and maize growing season in 2013 and 2014. (FS, fallow season; MS, maize growing season).

two rainfall events on September 18 (52 mm) and September 22 (40 mm) in 2013. This was probably due to the relatively low NO_3-N (2.6–8 mg kg^{-1}) and NH_4-N (3.8–8.1 mg kg^{-1}) contents, which limited the nitrification and denitrification rates [53, 58]. The cumulative N_2O emissions induced by rainfall accounted for 6.4 and 12.5% of the total annual N_2O emissions in 2013 and 2014, respectively.

Soil pH could also affect N cycling by directly affecting N mineralization and eventually N-containing gases [59]. Nitrogen mineralization linearly increased with soil pH, especially nitrification [60]. However, there was no significant relationship between pH and N mineralization in paddy waterlogged soils [61]. In calcareous soils with a high pH, ammonia volatilization was a loss pathway for fertilizer N and produced from soils under the condition of top dressing. Though the soil pH was 8.4 (calcareous soils), little ammonia volatilization appeared under the condition of buried fertilizer in this study [62].

There was a significantly positive correlation between N_2O emission and soil NO_3-N across the four N fertilization practices, which was consistent with previous studies [44, 46, 48], indicating that the soil mineral N was a key variable controlling N_2O emissions [22, 42], especially within 10 days after N fertilization. It was noted that N_2O emissions within 10 days after N fertilization increased linearly with soil NO_3-N content in both years 2013 and 2014 ($p < 0.05$). Liu et al. [42] also reported a linear relationship between N_2O emissions and soil mineral N content at the seasonal and annual scale. However, the slope was larger in 2014 (0.0093) than in 2013 (0.0058) (Fig. 7), probably due to the inter-annual variations in precipitation (523 mm in 2013 and 597 mm in 2014). The soil mean moisture within 10 days after N application was higher in 2014 (60%) than in 2013 (40%), which was conductive to the production of N_2O [8, 46, 63]. NI and SR also affected N_2O emissions by changing soil mineral N content. Figure 4 shows that the ammonium N content in all treatments reached a peak on April 27,

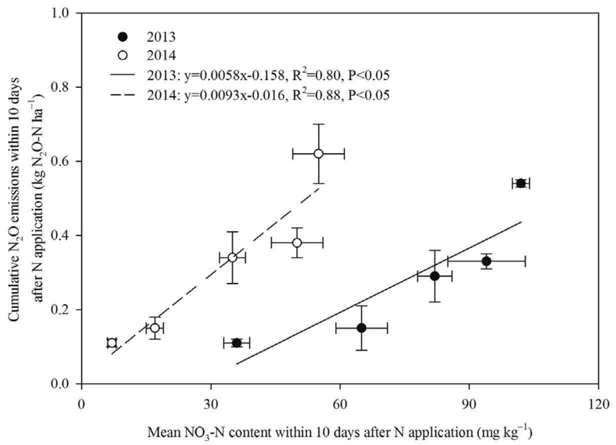

Figure 7. Relationship between mean soil NO_3-N at 0–20 cm depths and cumulative N_2O emissions within 10 days after N application.

2013, and it was higher in Opt + DCD treatment (45 mg kg^{-1}) than that in Opt treatment (42 mg kg^{-1}) because NI inhibited the first step of ammonium oxidation [31]. However, it was slightly lower in Opt + SR treatment (41 mg kg^{-1}), because SR fertilizers slowed down nutrient release rate through coating of the N fertilizers [11]. Thus, the highest nitrate N in Opt + DCD (98 mg kg^{-1}) and Opt + SR (151 mg kg^{-1}) treatments on May 23, 2013, was significantly lower compared with the Opt treatment (172 mg kg^{-1}) (Fig. 3). During the 2-year study period, the addition of NI significantly increased the ammonium N content and decreased the nitrate N content (Figs. 3 and 4). NI was effective in reducing N_2O emissions [8, 11, 64]. However, SR fertilizers had no such effect. In addition, Hu et al. found that the use of slow-release urea led to an increase in N_2O emissions (43%) during maize growing season and a decrease in N_2O emissions (33%) during wheat growing season [8]. This may be because high rainfall and irrigation during maize growing season accelerated SR hydrolysis [8], leading to N_2O emissions [9].

4.3 CH_4 uptakes and controlling factors

In this study, soils acted as a weak sink for atmospheric CH_4 by the oxidation of CH_4-oxidizing bacteria [7, 8, 45]. The annual CH_4 uptake ranged from 3.0 to 3.7 kgnCH_4-C ha^{-1} with an average of 3.4 kg CH_4-C ha^{-1}, which was significantly higher than that in an eastern corn belt Alfisols in the USA [65] and a corn field in Indonesia [64]. N fertilizer was regarded as an inhibitor to CH_4 consumption in soils [21]. However, it had no significant effect on CH_4 uptakes in this study, which was also reported by Shi et al. [19]. There was no significant relationship between soil temperature, WFPS, and CH_4 uptakes, but CH_4 uptakes tended to increase from April to July and decreased slowly in winter, which was in line with a previous study [46].

4.4 Implications for food production and greenhouse gas emissions in the semi-arid Loess Plateau

Overuse of N fertilizers is common in P. R. China. The three optimized N fertilization practices saved 20% of N fertilizer compared with the conventional one (200 kg N ha^{-1}), without decreasing grain yield. Shi et al. also found no significant decrease in grain yield with the reduction of N fertilizer from 200 to 185 or 186 kg N ha^{-1} y^{-1} in North China Plain [7]. After harvest, the nitrate accumulated in the 0–2 m soil profiles was reduced by 47.2, 48.5, and 45.5% in the three optimized N treatments as compared with the Con treatment, which minimized the risk of nitrate leaching.

The positive annual NGHG for all of the treatments (Tab. 1) indicated that N_2O emissions contributed significantly to the increase of NGHG in spite of CH_4 uptakes. Similar results were also reported for dry lands [7, 8, 66]. Compared to the Con treatment, NGHG was significantly reduced in the three optimized treatments due to their inhibitory effect on N_2O emissions. NGHGIs ranged from 26 to 78 kg CO_2-eq Mg^{-1} grain, which were comparable to that in maize systems in P. R. China (30–100 kg CO_2-eq Mg^{-1} grain) [8]. The largest decrease of NGHGI (48.7% in 2013 and 58.7% in 2014) was observed in Opt + DCD treatment, followed by Opt + SR treatment (43.6% in 2013 and 39.7% in 2014). These results indicated that Opt + DCD and Opt + SR treatments could significantly decrease N_2O emissions from soils while maintaining a high maize yield in the rain-fed regions of Loess Plateau. The spring maize is mainly planted in northern China with a plant area of seven million hectares [19], thus, the application of the most effective N fertilization practice, Opt + DCD, could save 0.35 million Mg N y^{-1} and 6.5 million kg N_2O-N y^{-1}.

The economic costs and benefits of these new N fertilization practices also need to be taken into account. The slow release urea used in this study was expensive relative to urea because of the complex manufacturing technology, and the NI used in this study was chemically pure, and thus, also expensive. Consequently, although NI has the potential to significantly decrease GHG emissions, farmers may be less willing to use it because of extra economic costs. A promising alternative solution is to add NI directly in the production of urea. In addition, subsidies or intervention policies from local government are also necessary for the application of these new N fertilization practices.

5 Concluding remarks

This 2-year field study investigated the effects of optimized N fertilization practices on GHG emissions in a rain-fed spring maize cropland. Soil acted as a small sink for atmospheric CH_4, and N fertilization practices had no significant effect on CH_4 uptakes. Soil acted as a source of N_2O, and N fertilization practices significantly influenced N_2O emissions. Soil temperature and moisture had no significant effects on N_2O emissions, but were closely related to fertilization and rainfall events (>40 mm). The background N_2O emissions and EFs in the rain-fed regions of Loess Plateau were much lower than the default values suggested by IPCC.

The three optimized N fertilization practices saved 20% of N fertilizer compared with the conventional treatment, but without a significant decrease in grain yield; and decreased the annual N_2O emissions, NGHG, and NGHGI. NI decreased cumulative N_2O emissions by about 72% within 10 days after N fertilization. Compared to the conventional treatment, NI significantly reduced annual N_2O emissions, NGHG, and NGHGI by 48, 56, and 54% on average, respectively. Moreover, NI decreased the accumulated nitrate in the 0–2 m soil profiles after harvest and minimized the risk of nitrate leaching. Therefore, Opt + DCD is the most effective strategy to mitigate N_2O emissions from the maize systems in the rain-fed regions of Loess Plateau. Reducing the cost of urea with NI was crucial for its widespread applications in agriculture.

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