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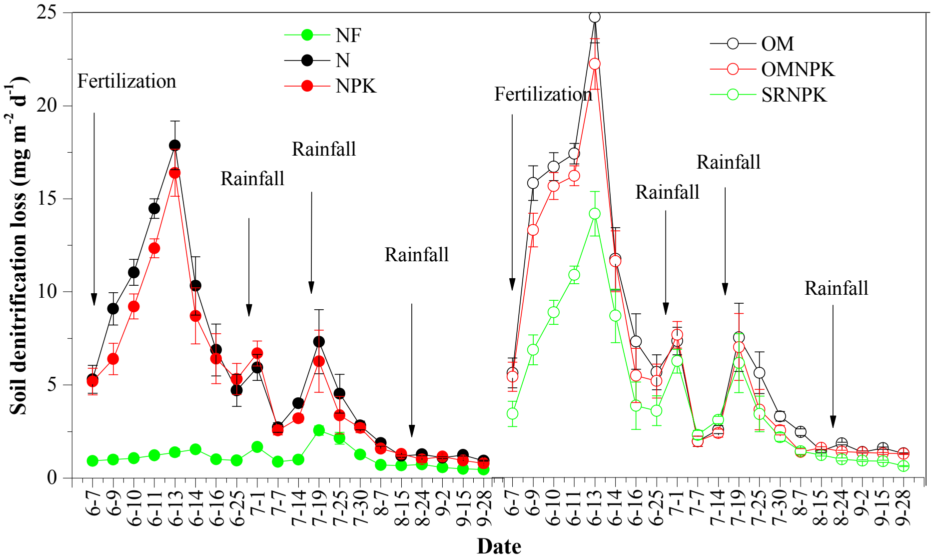

Environmental impact statement

Agricultural soils have been identified as responsible for approximately 60% of the total anthropogenic N_2O emissions. Nitrogen fertilizer application to agricultural soil will increase N_2O emission. Here we compared N_2O emissions and denitrification from rain-fed cropland under different fertilization regimes involved in single synthetic N fertilizer, organic manure, synthetic N, P, K fertilizer, organic manure with synthetic fertilizer, crop straw residue with synthetic fertilizer. Crop straw residue combined with synthetic fertilizer is recommended as an optimal strategy for mitigating N_2O emissions and denitrification-induced N loss.

The influence of N fertilization regimes on N_2O emissions and denitrification in rain-fed cropland during the rainy season

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Abstract The effects of nitrogen fertilization regimes on N_2O emissions and denitrification rates were evaluated by in situ field incubation experiments with intact soil cores and the acetylene block technique. Intact soil cores were collected from long-term field experiments involving several N fertilization regimes, including single synthetic N fertilizer (N), organic manure (OM), synthetic N, P, K fertilizer (NPK), organic manure with synthetic fertilizer (OMNPK), crop straw residue with synthetic fertilizer (SRNPK) and no nitrogen fertilizer (NF). N_2O was sampled from the head space of the cylinders to determine the daily N_2O emission and denitrification rate. The results showed that the N_2O emissions were greatly influenced by the specific fertilization regime even when the same nitrogen rate was applied. The mean N_2O emissions and denitrification rates from the N, OM, NPK, OMNPK and SRNPK treatments were 2.22, 2.66, 1.94, 2.53, 1.67 and 4.63, 5.96, 4.15, 5.41, 3.65 mg m^{-2} d^{-1}, respectively. The application of OM significantly increased the N_2O emission and denitrification compared to the application of NPK because of the high soil organic carbon (SOC) content of OM. However, SRNPK increased the SOC content and decreased the N_2O emissions significantly compared to the OM and OMNPK treatments because the addition of crop straw with a high C/N ratio to soil with a low inorganic N content induced N immobilization. The contents of soil nitrate and ammonium were the main limiting factors for N_2O emissions in a positive regression as follows: $\mathrm{Ln}\ (N_2O) = 2.511 + 1.258 \times \mathrm{Ln}\ ([NH_4^+] + [NO_3^-])$. Crop straw residue combined with synthetic fertilizer is recommended as an optimal strategy for mitigating N_2O emissions and denitrification-induced N loss in rain-fed cropland.

Keywords N_2O emission; Denitrification; Fertilization regimes; Soil inorganic nitrogen; Mitigation strategy; Rain-fed agriculture.

Introduction

As a potent greenhouse gas, nitrous oxide (N_2O) plays a key role with respect to global warming and climate change, with approximately 60% of global anthropogenic N_2O emissions originating from agricultural soils[1]. The magnitude of N_2O emissions from cropland is dependent on environmental factors as well as agricultural practices, such as tillage methods and N fertilizer rates and types[2, 3]. There are many reports on soil N_2O emission fluxes under different N rates and types[4, 5, 6, 7]. N_2O is an intermediate gaseous product of denitrification[8]. When different rates and types of N fertilizer are applied, the soil nitrate (NO_3^-) contents are undoubtedly changed. Several studies have demonstrated that the incorporation of organic matter, such as crop residues or livestock manure, could stimulate soil N_2O emissions[9, 10, 11]. In contrast, previous studies also demonstrated that the incorporation of high C/N ratio organic manure may result in a decrease in the soil N_2O emission during the denitrification process, particularly under conditions of high rainfall or irrigation[12, 13]. Lopez-Fernandez et al. demonstrated that the application of pig slurry reduced N_2O emissions by 27% compared to the application of urea in a Spanish xerofluent with a sandy loam soil[5]. Yao et al. reported that wheat straw application reduced N_2O emissions from a rice-wheat rotation field[14]. However, few reports on the effects of N_2O emissions under different types of N fertilizer were based on the comparison with the same N rates. It remains unclear whether this pattern may also apply to agricultural soils receiving the same amount of N but via different forms of N fertilizer. Meanwhile, the soil organic carbon (SOC) altered by an organic or inorganic fertilization regime may be another influencing factor on the soil N_2O emission[15, 16]. The contribution of denitrification to N_2O emissions from different levels of soil inorganic nitrogen and SOC derived from various fertilization regimes has not been well documented.

Rain-fed agriculture in southwestern China is an important agricultural practice because of the wet climate and poor irrigation conditions in the hillslope areas. Rain-fed cropland in the Sichuan Basin (the largest agricultural region in southwestern China) supplies 10% of the agricultural products of China[5], where Regosols (referred to locally as purple soil) are widely distributed over an area of 160,000 km^2 with dense populations and intensive agriculture during the rainy season[17]. Rain-fed agriculture in purple soil is an N-limited system with different practices of N amendments. There are some typical N-fertilization regimes, such as organic manure with pig slurry (OM), which has a long history of use; single synthetic N fertilizer (N), which was used in the 1970s; and synthetic nitrogen, phosphorus and potassium fertilizer (NPK) and OM plus synthetic NPK fertilizer and crop residues returned with synthetic NPK fertilizer, which are currently widely used. Therefore, gathering data on denitrification and its induced N_2O emission in rain-fed cropland during the rainy season is essential for evaluating N_2O pollution at the regional scale. N_2O emissions are 1.72 ± 0.34 kg N ha^{-1} yr^{-1} in rain-fed cropland due to the addition of N fertilizer to purple soil[2], but very few studies have investigated the impacts of fertilization regimes on N_2O emissions and the contribution of denitrification in rain-fed cropland with purple soil. In this study, we conducted in situ soil core incubation experiments using the acetylene block technique to examine the impacts of N fertilization regimes on N_2O production with the same rate of total N applied to rain-fed cropland with purple soil during the rainy season. We sought to determine a) the effect of the form of N fertilizer, i.e., synthetic fertilizer, manure, manure combined with synthetic fertilizer and crop residue combined with synthetic fertilizer, on N_2O emissions and the key parameters governing N_2O emissions; b) the contribution of denitrification to N_2O emissions; and c) the optimal fertilization regime for the mitigation of N_2O emissions.

Materials and methods

Field site

This study was conducted in Yanting Agro-Ecological Experimental Station of Purple Soil (N31°16′, E105°28′), Chinese Academy of Sciences, which is located in the central Sichuan Basin in southwestern China at an altitude of 400-600 m above sea level. The site is characterized by a moderate subtropical monsoon climate with an annual mean temperature of 17.3 ℃ and a mean precipitation of 826 mm, with rainfall mainly occurring during the summer and rainy season. The croplands are mostly rain-fed because of the topography, with slope gradients of 5-30% and a soil thickness of 30-80 cm, and typically lack an irrigation infrastructure[18].

Long-term fertilization experiments

A long-term fertilization experiment was established in 2003 with 5 fertilization treatments: single synthetic nitrogen fertilizer applied with NH_4HCO_3 (N); mixed synthetic nitrogen, phosphorus and potassium fertilizer (NPK); organic manure with pig slurry (OM); organic manure with pig slurry and synthetic fertilizer at a ratio of 40%:60% total N applied (OMNPK); and straw residue combined with synthetic fertilizer at a ratio of 15%:85% total N applied (SRNPK). The amount of N that was applied in both synthetic and organic forms was maintained at the same rate of 150 kg N ha^{-1} during the rainy season when maize (*Zea mays* L.) was planted and 130 kg N ha^{-1} during the dry season when wheat (*Triticum aestivum* L.) was planted in all of the fertilization treatments. Calcium superphosphate (90 kg P_2O_5 ha^{-1}) and potassium chloride (36 kg K_2O ha^{-1}) were applied to the NPK, OMNPK and SRNPK treatments, and a treatment without fertilizer (NF) was used as the control. Each of the treatments was replicated 3 times with an experimental plot area of 4 m × 8 m, each in a completely randomized design under conventional rain-fed cultivation. The plots were fertilized and sown with the late-maturing maize variety Zhongdan 808 on 7 June 2010. The field observation and incubation experiment spanned an entire rainy season, from 7 June to 28 September 2010. The main soil properties for the topsoil (0-15 cm) of the fertilization experiments in 2010 are listed in Table 1.

Table 1 Physical and chemical properties of the soil under different fertilization regimes in 2010.

Parameter	Fertilization regimes					
	NF	N	NPK	OM	OMNPK	SRNPK
pH	8.35^{a}	8.26 ^{b}	8.27^{b}	8.28^{b}	8.24^{b}	8.24^{b}
Bulk density (g/cm^3)	1.39^{a}	1.34^{b}	1.30^{b}	1.31^{b}	1.36^{b}	1.26^{c}
TN (g/kg)	0.25 ^{a}	0.35^{b}	0.56 ^{c}	0.88 ^{d}	0.79^{e}	0.73^{e}
SOC (g/kg)	4.24^{a}	4.51^{b}	6.43^{c}	9.89^{d}	8.63^{e}	9.96^{d}
C/N	16.96^{a}	12.89^{b}	11.48^{c}	11.24 ^{c}	10.92^{c}	13.64^{d}

Different letters in same line represent significant differences ($P < 0.05$) between treatments, according to Duncan test.

Measurements of the denitrification rate and N_2O emission

Eight intact soil cores (inner diameter 3.2 cm and length 18 cm) from each plot were placed into two different groups of incubation cylinders. All of the incubation cylinders were covered with caps with a small hole in the center of the top cap and sealed with a rubber stopper for gas sampling. Ten percent of the air from one group of cylinders was replaced with C_2H_2 to block the enzymatic reduction of N_2O to N_2 to determine the denitrification rate, while another group of cylinders without C_2H_2 was used to determine the natural N_2O emissions. Before each determination of denitrification rate, the same content of C_2H_2 (purified by bubbling through H_2SO_4 solution and distilled H_2O) had been injected into cylinder and then incubated for 24 h. The cylinders were sealed and buried for in situ incubation in the fertilization plots from which the soil cores were collected daily to measure the denitrification rate and natural N_2O emission. Gas samples of 10 ml were collected from the head space of the cylinders using a syringe for immediate N_2O analysis using a gas chromatograph (HP-5890 Series II, Hewlette Packard, Palo Alto, California, USA) equipped with an ECD. The N_2O emissions were measured daily immediately after fertilization, every other day after a rainfall event, which can trigger N_2O emissions, and once every week during the periods without rainfall. The daily mean emission for each treatment was calculated by averaging the 3 replicates for each measuring day. The total emissions for each treatment were calculated by integrating the daily losses with time.

The denitrification rate or N_2O emission was calculated as follows:

$$F = \frac{\frac{dCt}{1} \times Mw \times H \times 273.15 \times p \times 1000}{22.41 \times (273.15 + T) \times 1013}$$

[19],

where F is the daily denitrification rate (mg m^{-2} d^{-1} with C_2H_2) or N_2O emission (mg m^{-2} d^{-1} without C_2H_2), $\frac{dCt}{1}$ is the increment of N_2O concentration (ppbv) after 24 h of incubation, Mw (44 g mol^{-1}) is the molar weight of N_2O, H (m) is the height of the cylinder, p (10^5 Pa) is the atmospheric pressure, and T (℃) is the air temperature inside the cylinder.

The cumulative emissions of N_2O were calculated as follows:

$$E = \sum (E_{i+1}+E_i)/2 \times (t_{i+1}-t_i),$$

where E is the cumulative denitrification or N_2O emissions, i is the ith measurement, and the term $(t_{i+1}-t_i)$ represents the number of days between 2 consecutive measurements.

Measurements of environmental factors

During gas sampling, the soil temperature and volumetric water content at a 5-cm depth were measured, and soil samples from all of the plots were collected for NO_3^- and NH_4^+ analyses. The soil NO_3^- and NH_4^+ were extracted with 2 M KCl and determined using a continuous-flow analyzer (AA3, BRAN+LUEBBE, Germany). The measured water contents were converted into a water-filled pore space (WFPS) using the bulk density given above and a theoretical particle density of 2.65 g cm^{-3}[6]. The data for the daily precipitation, air pressure and temperature were obtained from the meteorological station at Yanting Agro-Ecological Experimental Station, which is 50 m away from the experimental site.

Data analysis and statistical methods

The significance of the differences in the N_2O emission, denitrification rate and soil properties between the different fertilization treatments was determined using a one-way ANOVA (SPSS Statistics Client 19.0, SPSS, Inc., USA). Pearson's test was used to determine the relationship between the N_2O emission, denitrification rate and soil properties. Graphical outputs were obtained from Origin 8.0 (Origin Lab Ltd., Guangzhou, China).

Results

Climatic and soil conditions

The air temperature was greater than 17.7 ℃ during the observation period (7 June to 28 September 2010). The mean air temperatures in June, July, August and September were 24.2, 26.8, 27.1 and 24.0 ℃ (Fig. 1a), respectively. The soil temperature at a 5-cm depth ranged between 19.4 and 33.1 ℃, with means of 26.4, 26.4, 25.0, 24.6, 25.2 and 24.6 ℃ in the NF, N, NPK, OM, OMNPK and SRNPK treatments (Fig. 1b), respectively. During the observed period, there were 55 days of rain and 12 days of heavy rain of more than 10 mm. The amounts of precipitation in June, July, August and September were 56.4 mm, 176.5 mm, 108.2 mm and 89.8 mm, respectively. Continual rain occurred from 23 to 28 June, with a rainfall amount of 11.4 mm; from 13 to 20 July, with 140.9 mm; and from 20 to 25 Aug, with 96.6 mm. The soil water-filled pore spaces (WFPS) ranged from 38.4%-81.6%. In general, the WFPS varied with the rainfall and temperature with over 65% of the WFPS after the three continual rain periods and below 65% for most of the days when the air temperature was greater than 26 ℃ (Fig. 1c). The SRNPK treatment had the lowest WFPS throughout most of the observation period.

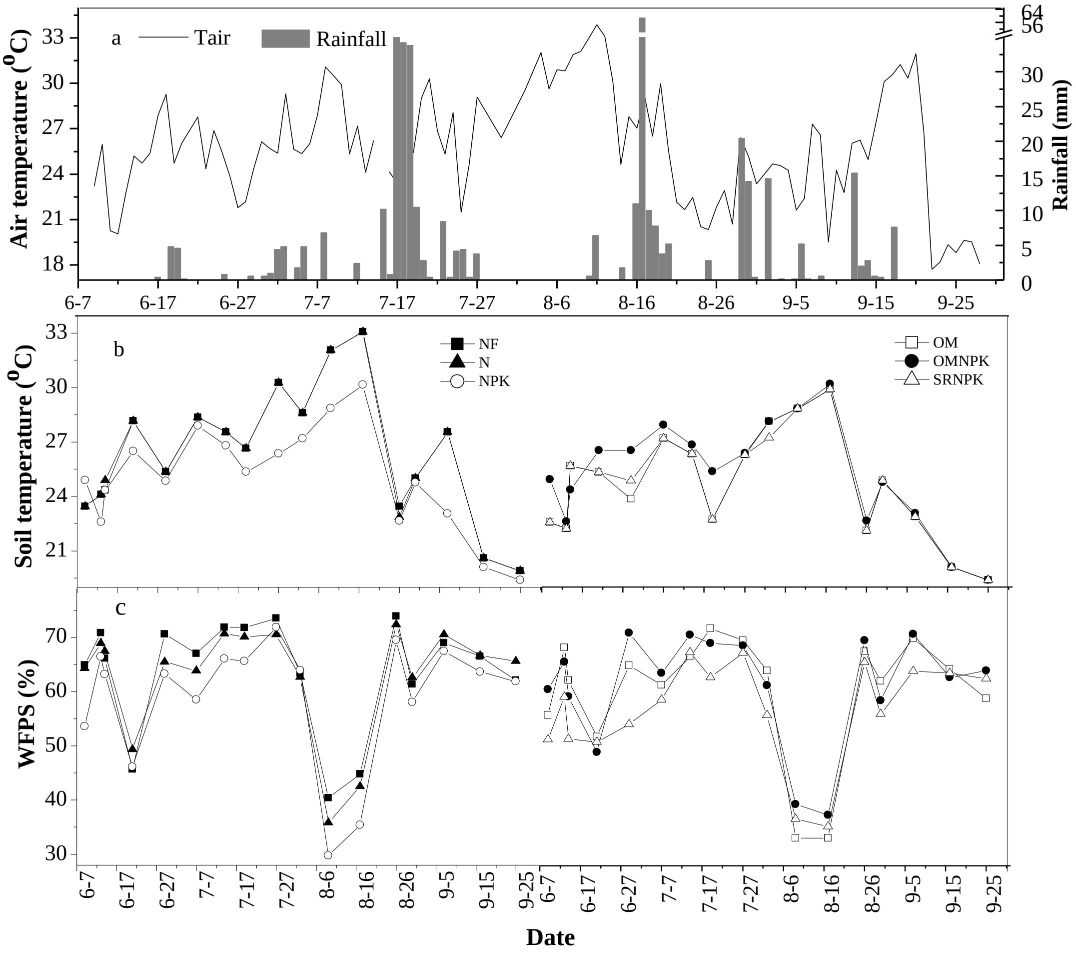

Fig. 1 Daily air temperature (℃), rainfall (mm), soil temperature (℃) and WFPS during the experimental period. (a) Daily air temperature (℃) and rainfall (mm); (b) soil (5 cm depth) temperature (℃); and (c) soil moisture content expressed as water-filled pore space (WFPS, %). The standard errors of the soil temperature and moisture are not shown for clarity.

The dynamic soil NH_4^+ and NO_3^- contents during the rainy season are shown in Fig. 2. The inorganic nitrogen content increased immediately after fertilizer application, and the highest soil NH_4^+ was observed 3 days following nitrogen fertilization, with values of up to 1.42-1.80 mg kg^{-1} in the N, OM, NPK and OMNPK treatments, and 7 days after fertilization in the SRNPK treatment, with a lower value of 1.39 mg kg^{-1} (no significant difference). The soil NH_4^+ decreased quickly and reached the lowest value (approximately 0.5 mg kg^{-1}) at approximately 2 weeks after fertilization (Fig. 2a). The soil NO_3^- peaked (9.90 - 45.25 mg kg^{-1}, $p<0.01$, $n=3$) 7 days after fertilization due to rapid nitrification and continuously decreased until maize harvest, with minimum values lower than 2.0 mg kg^{-1} (Fig. 2b), which can be ascribed to crop absorption, nitrate leaching and denitrification[18]. The OM and OMNPK treatments had significantly higher contents of NO_3^- and NH_4^+ than did the SRNPK treatment, especially during the first week following fertilization ($p<0.01$, $n=3$), whereas there were no significant differences among the N, NPK and OMNPK treatments (Fig. 2). Both the soil NH_4^+ and NO_3^- contents throughout the entire observation period followed the order OM>OMNPK>N>NPK>SRNPK>NF. Thus, organic manure provided more available N, and the inorganic N supplied from crop straw was relatively lower.

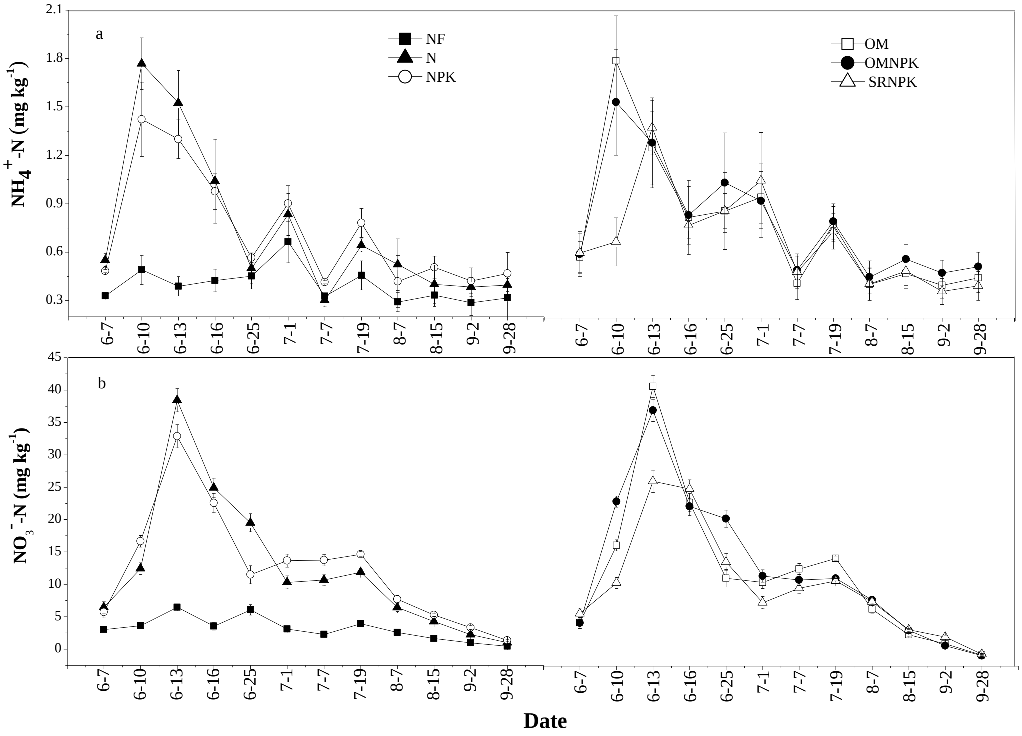

Fig. 2 Dynamics of the soil inorganic nitrogen contents during the rainy season. (a) Soil

ammonium content (mg kg^{-1}); (b) soil nitrate content (mg kg^{-1}). The vertical bars indicate the standard errors (SE) of the measurements from three replicated plots.

There were no significant differences in the pH among the different fertilizer treatments (Table 1). The long-term application of organic manure and crop straw led to significant increases in the SOC contents compared to the synthetic N, P and K fertilization ($p < 0.01$, n=6) (Table 1). The soil C/N ratio was greatest in the NF ($p<0.05$, n=6) treatment followed by the SRNPK treatment, whereas the OM and OMNPK treatments had the lowest C/N ratio (Table 1).

Soil N_2O emission

The N_2O emissions under different fertilization regimes are presented in Fig. 3. In general, the fertilizer application greatly stimulated the N_2O emissions during the first week after fertilization. The highest N_2O emission (8.71-13.33 mg m^{-2} d^{-1}) occurred on the sixth day after fertilization, and the differences in the N_2O emissions among the treatments were significant during the first week after fertilization ($p<0.01$, n=3). The OM and OMNPK treatments showed the highest N_2O emissions, with mean values of 10.58 and 9.35 mg m^{-2} d^{-1}, respectively, followed by the N and NPK (mean 6.94 and 4.62 mg m^{-2} d^{-1}, respectively) treatments. The SRNPK treatment had the lowest N_2O emission (3.73 mg m^{-2} d^{-1}) of all of the N fertilization treatments, and the N_2O emissions were also extremely low in the NF plots. The N_2O fluxes that were emitted within one week after fertilization comprised 13.8% to 20.7% of the total emissions during the entire maize-growing period (114 days). For the remaining maize season, the response of the N_2O emissions to different fertilization regimes was similar to that in the first week, but the differences among the treatments diminished (Fig. 3). The average N_2O emissions from the N, OM, NPK, OMNPK, SRNPK and NF treatments during the remaining period of the rainy season were 1.88 ± 0.49, 2.19 ± 0.54, 1.71 ± 0.44, 2.21 ± 0.59, 1.35 ± 0.34 and 0.53 ± 0.10 mg m^{-2} d^{-1}, respectively. Two N_2O emission peaks were captured due to heavy rainfall events (1 July and 19 July); thereafter, the N_2O emissions decreased continually with a low emission (less than 0.48 mg m^{-2} d^{-1}) and lasted until the end of the rainy season.

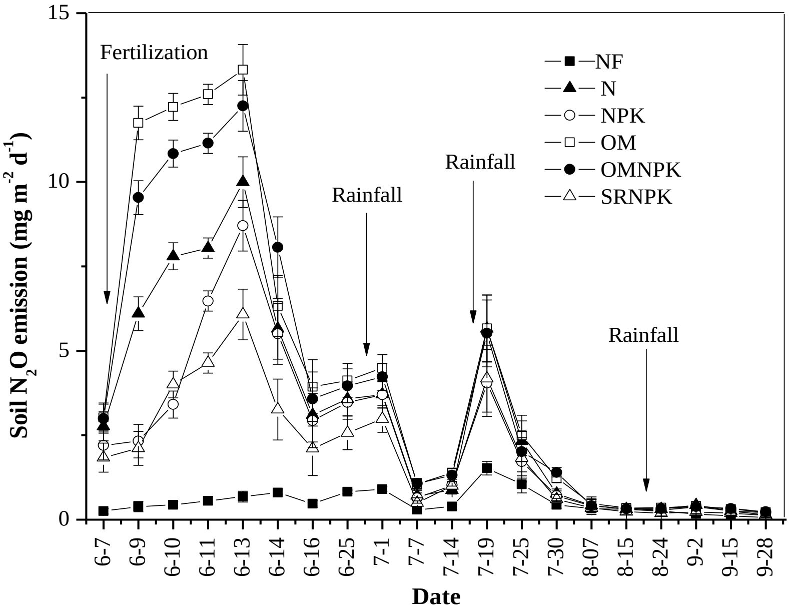

Fig. 3 Seasonal patterns of soil N_2O emissions during the rainy season under different fertilization regimes. The arrows represent the timing of the fertilizer applications and intense rainfall events. The vertical bars indicate the standard errors (SE) of measurements from three replicated plots.

Soil denitrification rate

The soil denitrification rates during the entire rainy season exhibited similar trends as were observed for the soil N_2O emissions but had higher values (Fig. 4). The soil denitrification rates exhibited sharp increases after fertilization or intense rainfall events in all of the treatments and varied from 0.94 to 17.87 mg m^{-2} d^{-1} in the N plots, 1.28 to 24.77 mg m^{-2} d^{-1} in the OM plots, 0.63 to 14.19 mg m^{-2} d^{-1} in the NPK plots, 1.31 to 22.24 mg m^{-2} d^{-1} in OMNPK plots and 0.79 to 16.40 mg m^{-2} d^{-1} in the SRNPK plots. The OM treatment showed the highest denitrification rate, with a mean of 5.96 mg m^{-2} d^{-1}, followed by the OMNPK (5.41 mg m^{-2} d^{-1}), N (4.63 mg m^{-2} d^{-1}), NPK (4.15 mg m^{-2} d^{-1}), SRNPK (3.65 mg m^{-2} d^{-1}) and NF (0.93 mg m^{-2} d^{-1}) treatments. The differences among the fertilization treatments were significant ($p<0.05$, n=3).

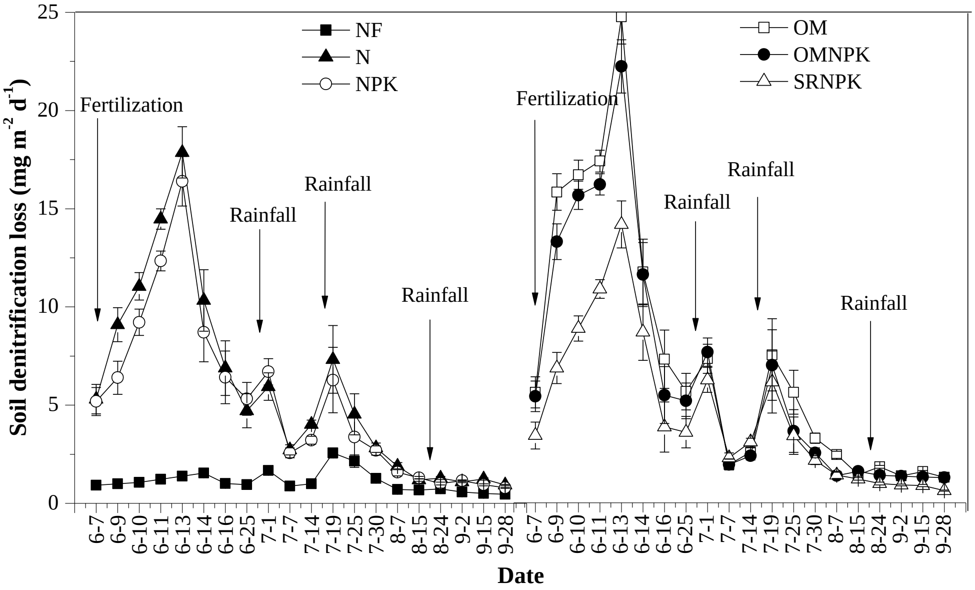

Fig. 4 Seasonal patterns of soil denitrification rates during the rainy season under different fertilization regimes. The vertical bars indicate the standard errors (SE) of the measurements from three replicated plots.

Cumulative nitrogen loss via soil N_2O emissions and denitrification

The cumulative N_2O emissions during the rainy season varied from 0.56 to 3.01 kg ha^{-1} among the fertilization regimes, which accounted for 0.88% to 1.63% of the applied N (Table 2). The accumulated nitrogen loss through denitrification ranged from 0.78 to 6.74 kg ha^{-1} in all of the fertilization treatments during the rainy season (Table 2), which accounted for 2.23%-3.97% of the applied N, respectively. The denitrification coefficients in the OM-related regimes (OM and OMNPK) were higher compared to conventional NPK fertilization and crop residue returned with synthetic fertilization (SRNPK) (Table 2). The seasonal cumulative denitrification-induced nitrogen losses among the fertilization regimes were significantly different ($p<0.05$), with the highest loss in the OM treatment, followed by the OMNPK, N, NPK, SRNPK and NF treatments. The ratio of cumulative natural N_2O emissions to denitrification was in the range of 0.45 to 0.48 in the N-fertilization treatments and the lowest in the OM treatment. However, the ratio was the highest in the NF treatment with significant differences from those in the N fertilization treatments ($p<0.01$) and (Table 2).

Table 2 Cumulative N loss through N_2O emissions and denitrification from the rainy season under different fertilization regimes.

Treatments	Cumulative N_2O emission (kg /ha)	Denitrification N loss (kg/ha)	N_2O emssion /denitrification	N_2O emission in applied N (%)	Denitrification coefficient (%)
NF	0.56^{a}	0.78^{e}	0.72^{a}	-	-
N	2.51^{bc}	5.23^{c}	0.48^{b}	1.30	2.97
OM	3.01^{d}	6.74^{a}	0.45^{cd}	1.63	3.97
NPK	2.19^{ce}	4.69^{d}	0.47^{bc}	1.09	2.61
OMNPK	2.86^{bd}	6.11^{b}	0.47^{bc}	1.53	3.55
SRNPK	1.88^{e}	4.12^{d}	0.46^{c}	0.88	2.23

Each value represents the mean of three replications; means followed by different letters are significantly different among fertilization treatments at $p \leq 0.05$.

Discussion

In the present study, the N_2O emissions exhibited the same trend as did the denitrification rates with high ratios of N_2O emissions to denitrification, meanwhile, the N_2O emission was significantly related to the denitrification rate (Table 3), indicating that denitrification was a major path of nitrogen loss from the rain-fed cropland in the Sichuan Basin. The pulse emissions of N_2O that were induced by heavy rainfall with WFPS greater than 65% were apparently the result of denitrification. The favorable WFPS levels for denitrification were generally in the range of 60%-90%[20-22]. Numerous studies have been published to demonstrate the role of the denitrification process in soil N_2O emission [6, 15, 22, 23].

Table 3 Correlations between the denitrification rate, N_2O emission and soil physical-chemical properties.

	SOC (g/kg)	TN (g/kg)	DOC (mg/kg)	C/N	NO_3^- (mg/kg)	NH_4^+ (mg/kg)	N_2O flux
Denitrification rate	0.379*	0.266	0.250	0.375*	0.854**	0.805**	0.991**
N_2O flux	0.329	0.203	0.226	0.369*	0.862 **	0.795**	

Available phosphorus, available potassium and bulk density had no significant correlations with N_2O emission, which were not shown in the table (n=30)

Denitrification is the stepwise reduction of NO_3^- to N_2 with an intermediate gaseous product of N_2O[24]. The application of nitrogen fertilizer in both organic and inorganic forms will increase the contents of soil nitrate and ammonium, which are substrates for denitrification and may increase N_2O emissions. A positive regression of the natural logarithm of N_2O or denitrification rate was significant compared to the logarithm of the corresponding soil NH_4^+ and NO_3^- contents (Fig. 5). This result is consistent with previous studies[15, 25, 26]. However, the cumulative N_2O emissions and denitrification differed greatly among the different fertilization regimes, even when the amount of nitrogen was applied at the same rate. Higher N_2O emissions were observed under long-term amendment with organic manure (Fig. 3, 4, Table 2). In addition to the soil inorganic nitrogen contents, the SOC was found positively correlated with the denitrification rate ($r=0.379$, $p<0.05$, $n=30$, Table 3). Kilian et al. and Chang et al. also observed that repeated applications of organic fertilizers resulted in increased SOC contents and N_2O emissions[7, 27]. The addition of organic manure increases denitrification because of the additional supply of reactive carbon[28, 29]. Soil organic carbon may trigger denitrification by enhancing respiration through the consumption of oxygen, which creates anoxic microsites, and by providing energy for denitrifiers[29]. The synthetic N, P and K fertilizer treatments showed lower cumulative N_2O emissions than the OM and OMNPK treatments due to short supplies of reactive carbon (Table 2, Fig. 2). Interestingly, long-term crop straw residues that were returned to cropland increased the SOC (Table 1) but did not increase the N_2O emissions, as observed in the organic manure treatments. However, this treatment (SRNPK) reduced the N_2O emissions by 60.1%, 52.1% and 13.9% compared to the OM, OMNPK and conventional synthetic NPK fertilization regimes, respectively. This result is consistent with Cai et al., who observed a 70% water- holding capacity (WHC) in a laboratory study with potato soil using the ^{15}N gas-flux method[30]. Zou et al. showed that wheat straw residue could slightly reduce the N_2O emissions from rice paddies[31], whereas Zhao et al. indicated that maize straw residue increased the N_2O

emissions from a wheat field[32]. Generally, straw residues with low C/N ratios may enhance N_2O emissions[33]. In contrast, high C/N ratios may reduce N_2O emissions because of temporary N immobilization, which delays the release of N into the soil[4, 34]. The incorporation of plant residues with C/N ratios greater than 18 may enhance N immobilization[35]. In this study, the returned crop residues with high C/N ratios (wheat straw has a C/N ratio of 35.7, and maize straw has a C/N ratio of 68.1) most likely stimulated the immobilization of the applied synthetic N fertilizer by soil microbes. This result is further supported by the observed decrease in inorganic N (NH_4^+ and NO_3^-) compared to the NPK treatments (Fig. 2), which might decrease the microbial denitrification activity and thus decrease the N_2O emissions[36]. However, organic manure provided more inorganic nitrogen than did crop straw residues (Fig. 2). The contents of soil nitrate and ammonium were the main limiting factors for the N_2O emissions and denitrification rate in the respective positive regressions as follows: $\mathrm{Ln}(N_2O) = 2.511 + 1.258 \times \mathrm{Ln}([NH_4^+] + [NO_3^-])$ and $\mathrm{Ln}(N_2O + N_2) = 0.963 + 0.951 \times \mathrm{Ln}([NH_4^+] + [NO_3^-])$ (Fig. 5). It is noteworthy that soil inorganic nitrogen was the main substrate of denitrification and might be regulated by soil C/N ratios. In soils with high NO_3^- contents, the SOC would be the main driver of N_2O production via denitrification, but the NO_3^- availability might control the denitrification rates in the low-N soil[37]. This result may somewhat explain the relatively high N_2O emissions for the OM and OMNPK treatments and the low N_2O emissions for the SRNPK treatment. Thus, the fertilization regime of crop straw residues combined with synthetic N, P and K fertilizers should be recommended for the mitigation of N_2O emissions and for the restoration of the soil SOC levels.

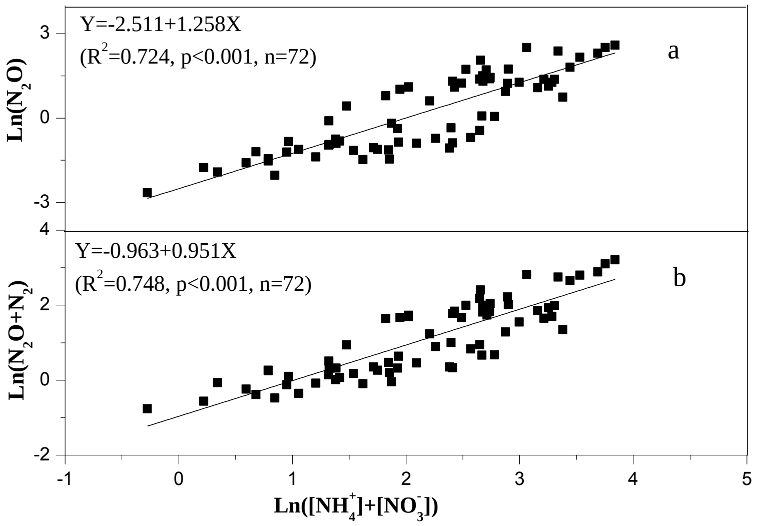

Fig. 5 Correlation of the soil inorganic nitrogen (NO_3^- +NH_4^+) contents with the N_2O emissions (a) and denitrification N loss (b).

The body of denitrification rate estimates, especially for terrestrial systems, is dominated by C_2H_2-based methods[38], and these estimates are reasonably robust, especially for systems with moderate or high NO_3^- levels[38]. In our study, fertilizer provided sufficient NO_3^- for most of the maize growing season. The results are reasonable when compared with the N_2O emissions as monitored by the static closed-chamber gas chromatograph method at the same field sites[1, 39]. The acetylene block technique also could effectively provide information for differences between the N_2O emission and denitrification rate among the fertilization regimes (Figs. 3, 4). However, this technique has been criticized due to precluding reliable estimates of total denitrification losses and has limitation when it is used to identify the N_2O sources[38,40,41], an accurate source of N_2O and the relative importance of denitrification and nitrification processes can be assessed in the future by isotope enrichment technique.

Conclusions

In this study, different fertilization regimes significantly affected the denitrification rates and N_2O emissions in a rain-fed cropland with purple soil during the rainy season. High N_2O emissions were triggered by N fertilization in the form of organic manure as a result of strong denitrification. The soil C/N ratio that was altered by the fertilization regime appears to regulate denitrification by altering the soil substrate NO_3^-, NH_4^+ and SOC contents. Crop straw residue returned with synthetic N, P and K fertilizer is recommended as the best nitrogen management practice for both N_2O mitigation and denitrification-induced N loss compared to the other widely used fertilization regimes of synthetic N, P and K, organic manure and organic manure with synthetic fertilizer at the same amount of applied nitrogen.

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