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Soil type affects not only magnitude but also thermal sensitivity of N₂O emissions in subtropical mountain area



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HIGHLIGHTS

- Annual N₂O emissions from acidic soil were significantly greater than for neutral and alkaline soils.
- Effects of soil pH on N₂O emissions were inconsistent between wheat and maize seasons.
- N₂O emissions for acidic soils were more sensitive to changing temperature.
- · Soil type affects both the magnitude and thermal sensitivity of N2O the emissions

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ABSTRACT

It is a concern whether the effect of soil type on N_2O emissions has to be considered for regional mitigation strategies and emission estimates in mountainous areas with inherent spatial heterogeneities of soil type. To date, there were few field experiments which investigated soil type effects on N₂O emissions. Thus a 2-year field study was conducted to measure N2O emissions and soil environmental variables from three different soils that were formed from similar parental rock under the same climate. Seasonal N2O fluxes ranged from 0.18 to 0.40 kg N ha⁻¹ for wheat seasons and 0.40 to 1.50 kg N ha⁻¹ for maize seasons across different experimental soils. The intra- and inter-annual variations in N₂O emissions were mainly triggered by temporal dynamics of soil temperature and moisture conditions. On average, seasonal N2O fluxes for acidic soils were significantly lower than for neutral and alkaline soils in cold-dry wheat seasons while significantly greater than for neutral and alkaline soils in warm-wet maize seasons. These determined differences of N₂O emissions were mainly caused by differences of initial soil properties across different soils. Moreover, seasonal N₂O fluxes were positively correlated with soil pH in wheat seasons, but negatively correlated in maize seasons. The temperature sensitivity coefficient (Q_{10}) of soil N₂O emissions for acidic soil (4.06) were significantly greater than those for neutral (1.82) and alkaline (1.15) soils. Overall, N₂O emissions for acidic soils were not only higher than those for neutral and alkaline soils but also more sensitive to changing temperature. The present study highlights that soil type is needed to be carefully considered for regional estimate and proposing mitigation strategy of N₂O emissions especially in subtropical mountain regions with inherent great heterogeneity of soil type.

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1. Introduction

N₂O emissions from agricultural soils account for approximately 60% of total global anthropogenic emissions (IPCC, 2014). However, these estimates have high uncertainties due to substantial spatial-temporal variations across different soil types, climates, and management practices (Shcherbak et al., 2014; Zhou et al., 2018; Wang et al., 2018). For example, previous studies have demonstrated that disregarding heterogeneities in soil type and the associated spatial disparities of N₂O emission may largely account for the uncertainties in regional and global N₂O emission estimates (Paustian et al., 2016). Recently, a global meta-analysis indicates that IPCC-Tier 1 estimates could lead to deviations of -75% and 35% for acidic and alkaline soils respectively when ignoring the soil pH effects on regional and global N₂O budgets (Wang et al., 2018). Although the scientific community began to evaluate effects of soil type on N₂O emissions but mainly through laboratory incubation experiment and short-term field experiment (Chirinda et al., 2010; Pelster et al., 2012), there were few multi-year field study to assess effects of soil type on N₂O emissions under same climate and agricultural management practice.

Soil N₂O is the intermediate product of soil biotic and abiotic N transformation processes that occur simultaneously in soils (Butterbach-Bahl et al., 2013; Heil et al., 2015). These N transformation processes and associated soil N₂O emissions are not only controlled by the availabilities of C and N substrates but also by other soil properties (Skiba and Ball, 2002; Butterbach-Bahl et al., 2013; Zhou et al., 2017b). For example, several previous studies have demonstrated that soil pH is the key factor of soil N₂O emissions (Bakken et al., 2012; Shaaban et al., 2018; Wang et al., 2018). Low soil pH generally decreases the activity of the nosZ genes (Shaaban et al., 2018) and constrains the assembly of functional N₂O reductase, thereby increasing the ratio of N₂O:N₂ during denitrification (Bakken et al., 2012). Likewise, previous field and laboratory studies also observed decreases in N2O emissions from forest and agricultural soils due to a pH increase after liming and/or biochar additions (Borken and Brumme, 1997; Obia et al., 2015). However, N₂O emissions may increase with an increase of soil pH because high pH can enhance the N₂O production processes of nitrification and dissimilatory nitrate reduction to ammonium (DNRA) (Stevens et al., 1998; Zhu et al., 2019). Thus, the response of N₂O emissions to soil pH remains uncertain.

In addition to soil pH, soil texture through regulating soil gas diffusivity and thereby the availability of oxygen (O_2) in soils could control soil N_2O production-consumption processes as well as diffusionemission processes (Gu et al., 2007; Butterbach-Bahl et al., 2013; Zhou et al., 2017b). For example, Mctaggart et al. (2002) found that N_2O flux was negatively correlated with soil gas diffusivity of Japanese upland soils, and Pilegaard (2013) reported that the optimal soil water-filled pore space (WFPS) for N_2O emissions from sandy loam soil is much higher than that for sandy clay loam soil.

Numerous studies have stated that soil moisture and temperature are key regulators of soil N₂O emissions (Butterbach-Bahl et al., 2013; Pilegaard, 2013). For example, Davidson et al. (1993) found that soil N₂O emissions have optimum soil moisture conditions of 70–80% WFPS. Soil temperature can affect soil N₂O emissions through regulating enzymatic activity, chemical nitrogen turnover rate, and gas diffusion process (Skiba and Ball, 2002; Butterbach-Bahl et al., 2013; Zhang et al., 2020). The temperature sensitivity (Q_{10}) of N_2O emissions is commonly defined as the factor by which N₂O emissions increase with a 10 °C rise in temperature (Schindlbacher et al., 2004; Zhou et al., 2018). However, soil substrate availability, microbial community composition and biomass, enzymatic activities, and soil porosity change with soil types (Rowlings et al., 2012; Gu et al., 2013), which might result in different Q10 values of N2O emissions in different soil types. Previous studies have reported Q10 values of N2O emissions even greater than the global mean Q₁₀ values of soil CO₂ emissions (mean: 3.0) (Schindlbacher et al., 2004; Parkin and Kaspar, 2006; Zhou et al.,

2018). Thus, direct quantification of the Q_{10} values of soil N_2O emissions via field measurements is necessary.

To improve the accuracy of N₂O emission estimates in the subtropical and tropical ecosystems, intensive field N₂O emission measurements have been conducted over the last decade (Liu et al., 2017b). However, the accuracy of regional and global N2O emission estimates still has been constrained likely by that soil type variability has previously been disregarded (Paustian et al., 2016), especially in the mountainous regions with great regional heterogeneities of soil type. To date, the multi-year field studies to investigate effects of soil type on N₂O emissions and the underlying mechanisms in subtropical agricultural soils are still limited. Therefore, we conducted a 2-year consecutive field study to simultaneously measure N2O emissions as well as soil environmental variables and agronomical performances from three different agricultural soils in a subtropical mountainous landscape under identical climate and agricultural management practice. The aim of this study was therefore to quantify the effect of soil type on N₂O emissions thereby exploring the main regulators. We expect to clarify the concern whether role of soil type in N₂O emissions was necessary to be considered when one proposes regional mitigation strategy and estimates regional emission of N₂O in mountainous area, where substantial spatial heterogeneity of soil type is the inherent property.

2. Materials and methods

2.1. Site description and experimental design

The field experiment was conducted at Yanting Agro-Ecological station of the Chinese Academy of Sciences (31°16′ N, 105°28′ E) in Southwest China. The climate is classified as a moderate subtropical monsoon climate with mean annual precipitation of 863 mm and mean annual air temperature of 17.3 °C over the last 3 decades (1990–2019).

The field experiment included three types of soil, i.e. acidic soil (AC, pH 5.09), neutral soil (NE, pH 6.75), and alkaline soil (AL, pH 8.37). The soils were all formulated from the similar parental bedrock of purplish sandstone (Table S1) and characterized by rapid weathering, i.e. the weathering and soil formulation process is mostly taken less than 50 years (Zhu et al., 2008). The three soils are classified as Eutric Regosol in accordance with the FAO Soil Classification. They are the main agricultural soil types in mountain area of the Upper Yangtze River watershed with distribution area of over 300,000 km² and feeding more than 10% of the Chinese population. For each soil type, three replicate field plots were established and randomly distributed. Each field experimental plot with a size of 7.5 m² (5 m \times 1.5 m) is hydrologically isolated with partition walls of cement. The partition walls were constructed at least 60 cm into the bedrock for avoiding the horizontal seepage flow to other plots. The chemical-physical properties of the topsoil (0-20 cm) are shown in Table 1.

All experimental plots were cultivated with the same winter wheatsummer maize rotation system and the same agricultural management practices since 2012. Briefly, ammonium carbonate (130 kg N ha⁻¹ for the wheat season and 150 kg N ha⁻¹ for the maize season), superphosphate (90 kg P_2O_5 ha⁻¹), and potassium chloride (36 kg K_2O ha⁻¹) were distributed and manually mixed into the surface soil layer (e.g., approximately 10–15 cm) in accordance with local management practices. The fertilizer application rates were based on local recommendations (Zhou et al., 2014). Winter wheat (*Triticum aestivum*) was sown in November and harvested in the following early May, and summer maize (*Zea mays* L) was sown with 0.45 m row spacing and 0.3 m plant spacing in late May and harvested in September.

2.2. Measurements of N₂O emissions

The soil N_2O emission measurements in the present study were conducted from October 2017 to September of 2019 (i.e. starting 5 years after the experimental plots had been established) by using the static

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Soil	ЬН	SOC	NL	C/N ratio	CEC	BD	Porosity	Clay	Silt	Sand	Available Fe	Available Mn
type		g/kg	g/kg		cmol(+)/kg	g/cm ³	%	%	%	%	mg/kg	mg/kg
AC	$5.09 \pm 0.11c$	$6.32\pm0.41a$	$0.87\pm0.01a$	$21.78 \pm 1.22b$	$2.48 \pm 0.09e$	$0.97 \pm 0.02b$	$59.40 \pm 0.75a$	$32.79 \pm 0.35a$	$50.52 \pm 1.24a$	$16.69 \pm 1.13b$	$0.51\pm0.05b$	$21.31 \pm 0.80b$
NE	$6.75\pm0.13b$	5.70 ± 0.29 a	$0.66\pm0.02b$	25.94 ± 1.27 a	$8.55\pm0.02c$	$1.17\pm0.03a$	$53.65\pm0.86b$	$17.86\pm0.68c$	53.10 ± 2.05 a	$29.04 \pm 1.61a$	$0.75\pm0.11a$	$22.92 \pm 0.43a$
AL	$8.37\pm0.01a$	5.80 ± 0.09 a	0.80 ± 0.03 a	$21.65\pm0.99b$	$8.22\pm0.09c$	$1.14\pm0.01a$	$53.89\pm0.73b$	$30.82\pm0.31\mathrm{b}$	$51.76\pm1.87a$	$17.42 \pm 2.13b$	$0.54\pm0.08b$	$6.45\pm0.84\mathrm{c}$
Different	letters within the s	ame column indica	te significant differe	ences among treatme	ints at $p < 0.05$ leve	el. AC, NE and AL ir	ndicate the acidic, ne	utral and alkaline so	il types, respectively			

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chamber-gas chromatography technique (Zheng et al., 2008; Zhou et al., 2014). Stainless steel square chamber bases ($50 \text{ cm} \times 50 \text{ cm}$) were preinstalled in the soil to a depth of 15 cm at each plot after basal fertilization and kept undisturbed throughout each cropping season. There was a groove (width and depth: 3 cm each) on top of each chamber base, used as water seal during chamber measurement. Stainless steel chambers (50 cm \times 50 cm \times 50 cm) were wrapped with a thermal insulation layer to minimize air temperature variations inside the chamber during the measurements. In addition, each chamber was equipped with two fans (10 cm diameter) to mix the headspace air, and a thermometer to measure the air temperature. To minimize the disturbance of experimental plots by measurement and sampling activities, a wooden walking board was setup at each plot before each cropping season.

The gas sampling was performed daily for the first week, and every other day for the second week following N fertilization, and then changed to twice per week throughout each cropping season. For each measurement, five gas samples were taken between 09:00 and 11:00 a.m., in 8-min intervals (0, 8, 16, 24, and 32 min after chamber closure) from the headspace of each chamber using 60-ml gas-tight syringes. All gas samples were analyzed within 8 h using a gas chromatograph (Agilent 7890-B, USA) equipped with an electron capture detector for N₂O analysis. The carrier gas flow, column oven temperature, and flame ionization detector were periodically calibrated based on the GC calibration protocol. Standards were run periodically throughout the sample run, i.e. for every 5 samples, the N₂O concentrations were calibrated using a reference gas with standard N₂O concentration (0.49ppmv, Chengdu Chenggang Messer Gas Products, Co., Ltd., Chengdu, China) (Parkin and Venterea, 2010). N₂O fluxes were calculated based on the change rate of N₂O concentration in the enclosed chamber headspace over time (correlation coefficients >0.95), as described by Zheng et al. (2008). The cumulative N₂O emissions were calculated by linear interpolation of the daily fluxes between gas sampling dates (Parkin and Venterea, 2010).

2.3. Auxiliary measurements

Soil samples were collected from each plot using a soil auger after each gas sampling, and the visible stones, roots, and other litters were removed manually before mixing completely. The mixed samples were then extracted with 2 M KCl solution (soil: solution =1:5 w/v), shaken for 1 h, and filtered through 0.45-µm polyethersulfone membrane (Whatman[@]) filter. The extracts were analyzed for NH₄⁺, NO₃⁻ and NO_2^- content using continuous flow analyzer (Auto Analyzer 3, SEAL Analytical, Germany). Soil dissolved organic carbon (DOC) was extracted with deionized water (soil: water =1:5 w/v), shaken for 1 h then centrifuged for 10 min at 4000 rpm. The supernatant was filtered through 0.45-µm polyethersulfone membrane (Whatman[@]) and analyzed using a continuous flow analyzer equipped with a chemical oxidation module (Auto Analyzer 3, SEAL Analytical, Germany). Soil clay mineral contents were measured by a multipurpose X-ray diffraction system (Ultima IV, Rigaku, Japan). Daily precipitation and air temperature were recorded by established automatic meteorological stations that is about 100 m from the experimental plots. The temperature and moisture content of the topsoil (0-5 cm) were monitored with a manual thermometer (JM624, Jinming Instrument Co. Ltd., Tianjin, China) and a portable frequency domain reflector probe (MP-406B, Zhongtian Precision Instruments Co. Ltd., Nantong, Jiangsu, China), respectively. The measured volumetric moisture was converted to WFPS (%) according to the following equation:

$$WFPS(\%) = V / [1 - (BD/2.65)] \times 100\%$$
(1)

where V is the soil volumetric moisture measured at each plot and BD is the soil bulk density.

The classic exponential regression equation was used to calculate the relationship between soil temperature and N₂O emissions, which was fitted according to Eq. (2), where E_{N2O} is soil N₂O emission rate, *a* and *b* are fitted parameters, and *T* is soil temperature.

$$E_{\rm N2O} = a \times \exp^{bl} \tag{2}$$

The Q₁₀ value was calculated by the following equation:

$$Q_{10} = \exp^{10 \times b} \tag{3}$$

After crop maturation, the whole plant biomass of wheat and maize was harvested from each plot and separated into root, straw, and grain parts, and then air-dried and weighed for subsequent crop yield calculation. A subsample of root, straw, and grain parts was prepared and oven-dried to constant weight at 75 °C then ground for total nitrogen content analysis with an elemental analyzer (Vario MACRO cube, Elementar, Analysensysteme GmbH, Langenselbold, Germany). Plant N uptake (kg N ha⁻¹) was calculated by the following equation:

$$\begin{aligned} Plant N uptake(kg N ha^{-1}) &= plant dry matter biomass \\ &\times plant N content \end{aligned} \tag{4}$$

and the yield-scaled N_2O emission intensity (g N Mg⁻¹) was calculated with the equation:

$$\begin{aligned} \text{Yield} &-\text{scaled } N_2 O \text{ emissions } \left(g \ N \ Mg^{-1}\right) \\ &= \text{seasonal fluxes/grain yield} \end{aligned} \tag{5}$$

2.4. Functional gene abundance

Soil samples for functional gene abundance analysis were taken following the maize harvest in September 2018, then stored in liquid nitrogen. The corresponding primer sequences are listed in Supplementary Table S2. Briefly, 0.5 g of fresh soil was extracted using the FastDNA spin kit according to the product instructions (MP Biomedicals, CA, USA). The quality of extracted DNA was analyzed using gel electrophoresis (0.8% agarose). The nitrification-related (ammonia oxidizing archaea (AOA) amoA, ammonia oxidizing bacteria (AOB) amoA, hao) and denitrification-related (narG, napA, nirS, nirK, norB, nosZ) functional genes were amplified with quantitative real-time polymerase chain reaction (PCR) (TIB8600, Triplex International Biosciences Co., Ltd., China). A standard curve was prepared with plasmid DNA from one representative clone containing each target gene. Each reaction mixture (16 μ l) included 10 μ l of 2 \times SYBR real-time PCR pre-mixture, 0.4 µl each of forward and reverse primers, and 1 µl of DNA template containing approximately 10 ng of DNA. PCR reactions were performed

according to the following program: 95 $^{\circ}$ C for 5 min for 40 cycles (95 $^{\circ}$ C for 15 s, 60 $^{\circ}$ C for 30 s).

2.5. Statistical analysis

All statistical analyses were performed using SPSS 24.0 software (SPSS Inc., USA). Soil properties, seasonal and annual N₂O fluxes, grain yield, plant N uptake, yield-scaled N₂O emissions, and functional gene abundance were tested for significant differences by the least significant difference at the p < 0.05 level with one-way ANOVA following the Tukey's multiple range test. Sources of variance were analyzed by univariate analysis. Significant differences among soils in the box-whisker plots were compared by the least significant difference at the p < 0.05level with one-way ANOVA followed by the post-hoc LSD test. The relationships between normalized N₂O fluxes and soil variables (NH₄⁺, NO₃⁻, NO_2^- , DOC, soil temperature, and WFPS) were explored using multiple linear regression analysis. Moreover, the relationships between daily N₂O emissions and soil temperature and between seasonal or annual N₂O fluxes and soil pH were explored using nonlinear regression. Figures were prepared using Origin 9.4 software (Origin Lab Corporation, Northampton, USA).

3. Results

3.1. Environmental conditions and crop productivity

Throughout the experimental period, average soil and air temperatures were 17.7 °C and 16.4 °C, respectively (Fig. 1a). The mean annual precipitation was 858.4 mm, while the precipitation during the maize growing season in 2019 (809 mm) was approximately twice as high as for 2018 (418 mm) (Fig. 1b). Soil WFPS was in the range of 7.49% to 67.11% (mean: 34.52%). There were no significant differences in soil temperature or moisture content between the different soils in either maize or wheat season (Fig. 2a-b).

The average soil NH₄⁺ concentrations varied significantly between the different soils in following order: AC (113.35 mg N kg⁻¹) > NE (54.48 mg N kg⁻¹) > AL (31.15 mg N kg⁻¹) (p < 0.05) (Fig. 2c). The mean soil NO₃⁻ concentrations for AC (28.6 mg N kg⁻¹) and AL (30.59 mg N kg⁻¹) soils were significantly greater than those for NE soil (20.09 mg N kg⁻¹) (p < 0.05) (Fig. 2d). Soil NO₃⁻ increased following fertilization then declined gradually, but with different patterns for different soils (Fig. 3b). The soil NO₂⁻ concentrations for AL soil (0.03–31.10 mg N kg⁻¹) were significantly higher than those for AC (0.01–2.5 mg N kg⁻¹) and NE (0.01–1.52 mg N kg⁻¹) soils (Fig. 2e). The average soil DOC concentrations for NE (45.70 mg C kg⁻¹) soil



Fig. 1. Temporal dynamics of (a) daily maximum (Max AT), minimum (Min AT) air temperature, daily precipitation and soil (10 cm) temperature of AC, NE and AL soils, and (b) water filled pore space (WFPS) of AC, NE and AL AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.



Fig. 2. Box-whisker plots of (a) soil temperature, (b) soil WFPS, (c) ammonium (NH $^+_4$), (d) nitrate (NO $^-_3$), (e) nitrite (NO $^-_2$), (f) dissolved organic carbon (DOC) concentrations and (g) nitrous oxide (N₂O) emission rate of different soil types in the wheat and maize seasons. In box-whisker plots, the top edges of the box indicate the 25th and 75th percentiles, the central line indicates the median, and the square white dots indicate the average, respectively. The maximum whisker lengths are specified as 1.5 times the interquartile range. Different letters mean significant differences among soils in the wheat and maize seasons (p < 0.05, one-way ANOVA, post-hoc LSD test). SDW is soil dry weight. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.

were significantly greater than those for AC ($30.62 \text{ mg C kg}^{-1}$) and AL ($32.03 \text{ mg C kg}^{-1}$) soils (Fig. 2f).

Crop yields and plant N uptake in the wheat and maize season were significantly affected by the soil type and year × soil type interaction (except for plant N uptake in the wheat season), however, no effects were detected for the year alone (Table 2). Crop yields were 2.57–3.32 Mg ha⁻¹ for the wheat season and 3.28–5.67 Mg ha⁻¹ for the maize season. NE soil showed the best yield performances, which were11.6%–59.8% and 1.2%–27.7% higher than those of AC and AL soils, respectively. The plant N uptake was 73.48–102.18 kg N ha⁻¹ for the wheat season and 96.68–140.22 kg N ha⁻¹ for the maize season. Compared to the 2018 maize season, plant N uptake was significantly higher by 41.55% in AC soil but lower by 14.75% in AL soil in the 2019 maize season (p < 0.05), respectively.

3.2. Soil N₂O emissions

Soil N₂O emissions exhibited significantly different dynamics between the different experimental soils (Figs. 3e and 4). Across different experimental years and soils, the soil N₂O emissions ranged from -4.35to 20.27 µg N m⁻² h⁻¹ for AC, -10.74 to 34.03 µg N m⁻² h⁻¹ for NE and -2.17 to 43.40 µg N m⁻² h⁻¹ for AL. The soil N₂O emissions were significantly higher in the maize season compared to the wheat season, which were in the range of 1.11 to 228.86 µg N m⁻² h⁻¹ for AC, -0.50 to 45.87 µg N m⁻² h⁻¹ for NE and -10.47 to 171.65 µg N m⁻² h⁻¹ for AL in the maize season. The N₂O emissions from AC, NE and AL soils peaked on average 35.5, 19.5, and 13.0 days after fertilization in the wheat season, and 32.5, 18.0, and 4.0 days in the maize season, respectively.



Fig. 3. Temporal dynamics of topsoil (0-15 cm) (a) ammonium (NH $^+_1$), (b) nitrate (NO $^-_3$), (c) nitrite (NO $^-_2$), (d) dissolved organic carbon (DOC) content and (e) soil nitrous oxide (N₂O) emission rates of different soil types for the wheat-maize rotation system. The data shown in all panels are average and standard error of four spatial replicates for each soil type. The black downward arrows indicate the fertilization date. SDW is soil dry weight. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.

 N_2O fluxes in the wheat season were significantly affected by the factors of soil type, year, and year \times soil type (p < 0.05), whereas in the maize season or on the annual scale the factors year or year \times soil type factor had no effect on N_2O fluxes (Table 2). Seasonal N_2O fluxes ranged from 0.18 to 0.40 kg N ha⁻¹ for the wheat season and 0.40 to 1.50 kg N ha⁻¹ for the maize season across all experimental soils. The average annual N_2O fluxes were 1.52 kg N ha⁻¹ for AC soil, 0.67 kg N ha⁻¹ for NE soil, and 0.78 kg N ha⁻¹ for AL soil. The N_2O emission pulses due to fertilization events significantly contributed to cumulative N_2O emissions in the maize season (81.84% for AC, 54.12% for NE, and 65.14% for AL); however, nonsignificant contributions of these pulses were observed during the wheat season (Fig. 3e). The maize season averagely accounted for 55.0%, 63.1% and 84.6% of annual N_2O fluxes for AL, NE and AC soils, respectively, throughout the experimental period (Fig. 4).

Yield-scaled N₂O emissions reflect both the crop yield and environmental performance of different soils with regard to N₂O emissions (Table 2). The annual yield-scaled N₂O emissions were in the range of 132.1–581.3 g N₂O-N Mg⁻¹ yield, across different years and soils. The average yield-scaled N₂O emissions for AC, NE, and AL soils, respectively, were 82.63, 77.00, and 119.00 g N₂O-N Mg⁻¹ yield for the wheat season and 346.96, 76.10, and 88.70 g N₂O-N Mg⁻¹ yield for the maize season. Compared with the wheat season, yield-scaled N₂O emissions of the maize season were on average 407.6% higher for the AC soil, 25.0% lower for the AL soil. Yield-scaled N₂O emissions of the NE soil showed inter-annual variability in the maize and wheat season.

3.3. Functional gene abundance

As shown in Fig. 5, there were significant differences in the abundance of N₂O-forming and N₂O-reducing functional genes among the three soils. Regarding the nitrification process, the abundance of functional genes ranged from 7.53×10^5 to 1.91×10^7 copies g^{-1} soil for the AOA *amoA* gene, 2.76×10^6 to 1.65×10^7 copies g^{-1} soil for the AOB *amoA* gene, and 1.38×10^8 to 5.05×10^8 copies g^{-1} soil for the *hao* gene, (Fig. 5a). The abundance of AOA *amoA*, AOB *amoA* and *hao* genes was significantly and positively correlated with soil pH, but not significantly affected by other soil properties (Table S5).

Nitrate-reducing microorganisms were evaluated by quantifying the abundance of *narG* and *napA* genes, which ranged from 5.88×10^5 to 7.17×10^5 copies g⁻¹ soil and 7.64×10^4 to 4.29×10^5 copies g⁻¹ soil across the experimental soils, respectively (Fig. 5b). No significant differences were observed in *narG* gene abundance among the three soils, whereas the gene abundance of *napA* was significantly and positively correlated with soil pH (Table S5). The nitrite-reducing bacteria functional gene abundances of *nirS* (3.01×10^5 to 1.09×10^6 copies g⁻¹ soil) and *nirK* (5.14×10^3 to 4.22×10^4 copies g⁻¹ soil) showed significant differences (p < 0.05) among the soils, i.e., AL > NE > AC. Gene copies of the nitrous oxide reducing functional gene *nosZ* were lowest in AC soil (7.91×10^4 copies g⁻¹ soil), followed by AL soil (1.81×10^5 copies g⁻¹ soil) and NE soil (5.91×10^5 copies g⁻¹ soil).

3.4. Correlations between soil N₂O emissions and environmental factors

Soil N₂O emissions were mainly affected by soil temperature, WFPS, mineral N substrate (NH_4^+ , NO_3^- , NO_2^-), and DOC content across the different experimental years and soils (Table S3). The linear regression model of these environmental factors explains daily N₂O emission variations of 9.0%–15.0% in the wheat season, 27.0%–51.0% in the maize season, and 26.0%–49.0% on the annual scale (Table 3). Across the three soils, the exponential relationship between N₂O fluxes and soil pH was

Soil type	Wheat season				Maize season				Annual			
	N ₂ O flux	Grain yield	Plant N uptake	Yield-scaled N ₂ O	N ₂ O flux	Grain yield	Plant N uptake	Yield-scaled N ₂ O	N ₂ O flux	Grain yield	Plant N uptake	Yield-scaled N ₂ O
	$(kg N ha^{-1})$	$(Mg ha^{-1})$	$(kg N ha^{-1})$	(g N ₂ O-N Mg ⁻¹)	$(\mathrm{kg}~\mathrm{N}~\mathrm{ha}^{-1})$	$(Mg ha^{-1})$	$(\mathrm{kg}~\mathrm{N}~\mathrm{ha}^{-1})$	$(g N_2 O-N Mg^{-1})$	(kg N ha^{-1})	$(Mg ha^{-1})$	$(kg N ha^{-1})$	$(g N_2 O-N Mg^{-1})$
2018 AC	0.24 + 0.01h	2.6 ± 0.1h	735 + 76h	91 4 + 5 5a	1 50 ± 0 25a	45 U 3h	991 + 5 Ar	463 8 + 87 1a	1 74 + 0 76 a	5.6 + 0.4h	175 5 ± 13 9h	581 3 + 96 8a
NE	$0.18 \pm 0.01c$	$3.3 \pm 0.1a$	$100.0 \pm 16.1a$	$54.5 \pm 3.8b$	$0.41 \pm 0.02b$	$5.2 \pm 0.3a$	$131.1 \pm 6.5a$	$77.4 \pm 0.9b$	$0.59 \pm 0.03 \mathrm{b}$	8.6 ± 0.2 a	$243.8 \pm 10.3a$	$132.1 \pm 3.3b$
AL	0.30 ± 0.01 a	$3.1\pm0.1a$	$90.4\pm6.2a$	$98.9\pm5.4a$	$0.40\pm0.02b$	$5.2\pm0.2a$	$113.4 \pm 4.7b$	77.2 ± 1.6b	0.70 ± 0.02 b	$8.3\pm0.3a$	$212.0\pm6.1ab$	$176.0 \pm 4.1b$
2019												
AC	$0.22\pm0.03c$	$2.9 \pm 0.1b$	$89.2 \pm 3.6b$	$73.9\pm6.1c$	1.08 ± 0.09 a	$4.7\pm0.1b$	$140.2 \pm 3.9a$	$230.1\pm21.6a$	1.30 ± 0.07 a	$7.7 \pm 0.1b$	253.8 ± 5.5 a	$301.0\pm17.0a$
NE	$0.32\pm0.02b$	3.3 ± 0.04 a	$102.2 \pm 5.5a$	$99.5\pm4.0\mathrm{b}$	$0.42\pm0.04\mathrm{b}$	5.7 ± 0.04 a	$132.3 \pm 3.6b$	$74.8 \pm 7.0 \mathrm{c}$	$0.75\pm0.04~\mathrm{b}$	$8.9\pm0.1a$	$250.2 \pm 3.5a$	$174.3 \pm 9.4c$
AL	$0.40\pm0.06a$	$2.9\pm0.03b$	99.0 ± 2.9a	$139.1\pm12.1a$	$0.45\pm0.03b$	$4.4\pm0.1c$	$96.7 \pm 4.5c$	$100.2 \pm 3.8b$	$0.85\pm0.03~{ m b}$	$7.3 \pm 0.1b$	$225.5\pm8.2b$	$239.3 \pm 10.2b$
Source of varia	JCe											
Year	**	NS	NS	**	NS	NS	NS	**	NS	*	**	NS
Soil type	**	**	*	**	**	**	**	**	**	**	**	**
Year×soil type	**	NS	**	NS	**	* *	*	**	NS	**	**	**

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Table 2

found positive (p < 0.01) in the wheat season while negative in the maize season or on the year-round scale. (Fig. 7). Soil N₂O emissions increased exponentially with soil temperature in the three soils, exhibiting Q₁₀ values of 4.06, 1.82, and 1.15 for AC, NE, and AL soils, respectively (Fig. 6).

4. Discussion

4.1. Soil type and N₂O emissions

In the present study, the average seasonal N₂O fluxes were 0.28 kg N ha^{-1} season⁻¹ for wheat and 0.71 kg N ha^{-1} season⁻¹ for maize (Table 2). Our observations fall within the range of previously reported N₂O emissions for wheat and maize systems whereas the mean values are both lower than the global averages of 1.44 kg N ha⁻¹ season⁻ and 3.01 kg N ha⁻¹ season⁻¹ for wheat and maize systems, respectively (Linguist et al., 2012). In addition, the intra- and inter-annual dynamics of soil N_2O emissions were also observed in this study (Fig. 3, Table 2). although under same agricultural management practices and environmental conditions. Previous studies have effectively documented that the dynamics of soil environmental variables are the main drivers of soil N₂O emissions variation in various ecosystems (Luo et al., 2012; Rowlings et al., 2012; Zhou et al., 2017a). In this study, soil moisture conditions (WPFS) were positively correlated with N₂O emissions (Table 3), suggesting that rainfall dynamics likely contribute to temporal variations of N2O emissions. Nevertheless, the current study indicates that rainfall events do not always induce high pulses of N₂O emissions (Fig. 3), which contrasts with previous findings of rainfall events as triggers of hot moments of N₂O emission (Yao et al., 2015; Song et al., 2019; Shang et al., 2020). Pulses of soil N₂O emissions were only observed after large rainfall events in combination with N fertilization events in maize seasons and not after rainfall events in wheat seasons (Fig. 3). The lack of N₂O pulses after rainfall events in the wheat season was most likely due to nitrification-denitrification processes being hindered by low soil temperature (Szukics et al., 2010; Butterbach-Bahl et al., 2013). The absence of soil N₂O emission pulses after rainfall events during July to September (maize season) could be explained with NO_3^- limitation (Table 2, Fig. 3) because high $NO_3^$ leaching loss often occurs after large rainfall events thereby decreasing soil NO₃ availability for further N₂O production and emission (Zhou et al., 2012; Zhou et al., 2013).

Although there were intra- and inter-annual variations in soil N₂O emissions, the soil N₂O flux was significantly affected by soil types throughout the experimental period (Table 2). Seasonal cumulative N₂O fluxes from AL were significantly greater than those for AC and NE in wheat seasons, whereas seasonal cumulative N₂O fluxes for AC were significantly greater than for NE and AL in maize seasons. The different soil properties (e.g., pH, clay content, and porosity, Table 1) may have contributed to the significant effects of soil type on N₂O emissions (e.g., reviewed by Butterbach-Bahl et al., 2013; Heil et al., 2015).

As soil pH has been shown a key modifier of both biotic and abiotic N transformation processes related to N₂O production and consumption (Butterbach-Bahl et al., 2013; Heil et al., 2015; Wang et al., 2018), the differences in N₂O emissions for the three experimental soils in this study may be related to soil pH. The two years of consecutive observations indicate that the magnitude of N₂O emissions is positively correlated with pH, i.e. that soil N₂O emissions are higher at higher soil pH in the relatively dry and cold wheat season (Table 2). This phenomenon could be explained by the following reasons. First, compared to acidic soils, alkaline soils favor rapid formation of free NH₃, thereby directly providing substrates of ammonia oxidation processes for N₂O production (Kowalchuk and Stephen, 2001; Butterbach-Bahl et al., 2013). Second, greater nitrifier functional gene (AOA amoA, AOB amoA, and hao) abundance (Fig. 5) and 2:1 type clay mineral contents (Tables S4 and S6) in the neutral and alkaline soils could biologically and physically enhance nitrification rates, thereby increasing soil N₂O production in



Fig. 4. Temporal cumulative N_2O emissions (kg N ha⁻¹) from the different soils during 2017–2018 (a) and 2018–2019 (b). The data shown in all panels are average and standard error of four spatial replicates for each soil type. The black downward arrows indicate the fertilization date. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.

particular under the aerobic conditions of dry wheat seasons (Butterbach-Bahl et al., 2013; Paustian et al., 2016). Third, N₂O is mainly derived from nitrification at well-aerated, i.e., relatively dry soil conditions (Duan et al., 2018). Similar to our findings, Khan et al. (2011) also detected greater N₂O emissions at higher soil pH from a Paparua Templeton silt loam soil under dry conditions, which was most likely because limitation of N₂O production by low soil moisture conditions was faster and stronger in acidic soils than in neutral or alkaline soils. In contrast, in wet and warm maize seasons, the current study indicates that soil N₂O emissions from acidic soil tends to be greater than other soil types (Table 2), which is in line with a recent global meta-analysis of soil pH effects on N₂O emissions from agricultural soils (Wang et al., 2018). Some previous studies have also reported negative correlations between soil N₂O emissions and soil pH, i.e. with low pH tending to increase soil N₂O emissions (Liu et al., 2017b; Wang et al., 2018; Shaaban et al., 2018; Wu et al., 2018) (Fig. 4, Fig. 7). For example, the pulses of N₂O emissions following fertilization events in acidic soils were consistently more than two weeks longer than those in high pH soils throughout the whole experimental period (Fig. 3e). These results could be explained by the following mechanisms. First, in warm and wet maize seasons, the average soil NH⁺₄ contents for acidic soils (AC treatment: 59.3 mg N kg⁻¹) were significantly higher than those for high pH soils (NE treatment: 12.5 mg N kg⁻¹, AL treatment: 2.5 mg N kg⁻¹) following fertilization events, which in turn increased the availability of soil NH₄⁺ for nitrification processes (Figs. 2 and 3). Second, the relatively slow nitrification of acidic soil not only generates N₂O directly but also continuously produces NO₃⁻ and increases soil NO₃⁻ availability (mean: 42.3 mg N kg⁻¹ for AC, 21.5 mg N kg⁻¹ for NE, and 11.8 mg N kg⁻¹ for AL), thereby enhancing the denitrification potential, especially in combination with rainfall events (e.g., Butterbach-Bahl et al., 2013). Third, the low nosZ gene abundance of AC suggests a decreased activity of N₂O-reductase, and therefore an inhibition of complete denitrification. This will lead to a concomitant increase in the denitrification $N_2O/(N_2O + N_2)$ production ratio, and thus to an increase in soil N_2O emissions (Wu et al., 2018; Shaaban et al., 2018). Soil N₂O emissions are dependent on various biotic and abiotic N transformation processes that relate to the production and consumption of N₂O in soils, however, the regulating mechanisms of soil pH in these processes are still unclear (Jiang et al., 2015; Liu et al., 2017a), suggesting the need for further targeted study.



Fig. 5. Functional gene abundance (copies g^{-1} SDW) in the topsoil (0–15 cm) for the nitrification processes (a) (AOA *amoA*, AOB *amoA*, hao) and for the denitrification processes (b) (*narG*, *napA*, *nirS*, *nirK and nosZ*). Vertical bars indicate standard errors of four replicates. Different letters denote significant differences between soil types (p < 0.05). SDW is soil dry weight. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.

In addition to soil pH, soil texture could moderate soil O₂ availability and regulate soil N₂O emissions by impacting the size and distribution of soil pores (Groffman and Tiedje, 1991; Corre et al., 1999; Butterbach-Bahl et al., 2013; Zhou et al., 2017a; Song et al., 2018). In this context, differences in soil texture may be also another explanation for the observed effects of soil type (Tables 1 and 2). Specifically, it is possible that the different clay contents of different soils contributed to the significant effects of soil type on annual N₂O fluxes. This is likely because higher clay particles hold water tightly in soil aggregates and have low gas diffusivity, which might raise the potential for formation of anaerobic microsites, thereby favoring soil N₂O emission from denitrification (Gu et al., 2013). Low soil N₂O emissions at high clay content (> 40%) have often been observed in previous studies, most likely due to low gas diffusivity, which promotes complete denitrification with reduction of N₂O to N₂ (Weitz et al., 2001). Similarly, some studies have highlighted that soil gas diffusivity is a predictable indicator of soil N₂O emission potential (Balaine et al., 2013).

4.2. Temperature sensitivity of N₂O emission

In the present study, soil N₂O emissions increased exponentially with soil temperature in all three soils (Fig. 6), which is consistent with previous field or laboratory studies on various ecosystems (Yao et al., 2010; Song et al., 2018; Zhou et al., 2018; Li et al., 2020). The temperature sensitivity of soil N₂O emission varied substantially among soils, with the Q₁₀ values of soil N₂O emissions ranging from 1.15 to 4.06, which are comparable with values obtained in previous field studies (Smith et al., 1997: 1.5-5.0), as well as in incubation studies (Song et al., 2018: 1.1–5.3). Given the dominance of biological enzymes that catalyze the production of N₂O (Butterbach-Bahl et al., 2013), the temperature dependence of N₂O emissions is understandable because the enzymatic activity generally increases with rising temperature within an appropriate range, as long as no other factors limitations exist. Moreover, the increasing temperature could also physically enhance soil N₂O diffusion and emission processes (Gu et al., 2013; Butterbach-Bahl et al., 2013).

The present study showed that the Q_{10} values of N_2O emissions increased significantly with decreasing soil pH (Fig. S1), which is consistent with recent findings in steppe systems (Zhang et al., 2020). This pattern could be related to the effects of soil pH on the activities of N_2O -generating processes and on the form and availability of the substrate, which then directly/indirectly affecting the Q_{10} of N_2O emissions (Yao et al., 2010; Song et al., 2018). Moreover, it also could be related to a stronger shift from biological N_2O production with lower temperature sensitivity at higher soil pH to more abiotic (chemical) N_2O production with high temperature sensitivity at lower soil pH (Liu et al., 2017a; Liu et al., 2019). The activation energy of specific substrates for N_2O production is relatively stable for the given N_2O production pathways (Butterbach-Bahl et al., 2013; Heil et al., 2015). Thus, the variability of Q_{10} would be decided by the relative proportions as well as the



Fig. 6. Relationships between N₂O emissions and topsoil temperature for the AC, NE and AL soils throughout the experimental periods of 2017–2019. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.

magnitudes of different N₂O production pathways for different soils (Blagodatskaya et al., 2014; Song et al., 2018). Some studies have shown that soil pH may affect the microbial communities and subsequent biogeochemical processes of N₂O production and consumption (Fierer and Jackson, 2006; Nicol et al., 2008). For example, Nicol et al. (2008) and Jiang et al. (2015) reported that the soil pH has an important role in shaping the community structures and abundance of ammonia oxidizers in upland and paddy soils, respectively. Similarly, the current study found that the abundance of functional genes (AOA *amoA*, AOB *amoA*, *hao*, *napA*, *nirS*, *and nirK*) in soil N₂O emissions is correlated

Table 3

Results of linear regression analysis of the effects of soil moisture (WFPS, in %), soil temperature (ST, in $^{\circ}$ C), mineral nitrogen substrate (MN=NH₄⁺+NO₃⁻ + NO₂⁻) and soil DOC content on N₂O emissions from the AC, NE and AL soil throughout the experimental periods of 2017–2019.

Season	Soil type	Regression function	R ^{2a}	p^{b}	n ^c
	AC	$Ln(N_2O) = 0.33ST + 0.24WFPS + 0.20Ln(MN)-0.15Ln(DOC)-0.66$	0.15	< 0.05	185
Wheat season	NE	$Ln(N_2O) = 0.27Ln(MN) + 0.15WFPS + 1.96$	0.09	< 0.05	193
	AL	$Ln(N_2O) = 0.37Ln(MN) + 1.22$	0.13	< 0.001	176
	AC	$Ln(N_2O) = 0.49Ln(MN) + 0.33WFPS + 0.29ST - 2.74$	0.35	< 0.001	212
Maize season	NE	$Ln(N_2O) = 0.47Ln(MN) + 0.39WFPS + 0.20ST-1.16$	0.27	< 0.001	208
	AL	$Ln(N_2O) = 0.77Ln(MN) + 0.25WFPS - 1.63$	0.51	< 0.001	202
	AC	$Ln(N_2O) = 0.72ST + 0.33Ln(MN) + 0.28WFPS - 3.08$	0.49	< 0.001	391
Year-round	NE	$Ln(N_2O) = 0.50ST + 0.30WFPS + 0.22Ln(MN) - 0.16Ln(DOC) + 0.29$	0.26	< 0.001	382
	AL	$Ln(N_2O) = 0.62Ln(MN) + 0.22ST + 0.20WFPS-1.27$	0.34	< 0.001	391

^a Coefficient of determination.

^b Level of significance.

^c The symbol "n" represents the number of measurements. AC, NE and AL indicate the acidic, neutral and alkaline soil types, respectively.



Fig. 7. Relationships between soil pH and cumulative N_2O [wheat season (a), maize season (b) and annual (c)] emissions from 2017 to 2019.

with soil pH (Table S5). In addition, the relatively low but long-lasting nitrification process at low pH soils (i.e., AC soil) not only directly generates N_2O but also produces NO_3^- , thereby increasing soil NO_3^- availability for denitrification and N_2O emissions (Fig. 2). The structural equation model (SEM) analysis explained 95% of the variance in Q_{10} values (Fig. S1), which consequently indicates that soil pH, AOB *amoA* gene abundance, and clay minerals could be the key controlling factors for the thermal sensitivity of N_2O emissions.

4.3. Implications and perspectives

A better understanding of the effects of soil type on N_2O emissions via multi-year field measurements can contribute to developing mitigation strategies and improving the accuracy of regional and global N_2O inventories. For example, the acidic soil has been found to be a "hot spot" of N_2O emissions in the summer-maize season due to the warm and wet conditions as compared with the other two soils. Thus, our findings highlight that pH management is crucial for mitigating soil N_2O emissions, in particular for tropical and subtropical regions with widespread acidic soils and wet and warm conditions. For alkaline and neutral soils, the application of nitrification inhibitors could inhibit the ammonia-oxidizing process and reduce N_2O emissions (Tian et al., 2017; Wu et al., 2017), representing an optimal management practice for mitigating soil N_2O emissions in these soils (Tierling and Kuhlmann, 2018). In addition, soil types not only control the dynamics and magnitude of N₂O emissions but also regulate the thermal sensitivity (Q_{10}) of soil N₂O emissions. For example, a recent global meta-analysis reported a negative relationship between N₂O emissions and soil pH (Wang et al., 2018), this contrasts with the response pattern of N₂O emissions to soil pH observed in this study, which is likely due to different Q₁₀ values among different soils. Thus, soil properties (especially soil pH) should be carefully considered when estimating regional and global N₂O emissions and predicting the response of soil N₂O emissions to climate warming.

5. Conclusion

The results of the present study indicate that soil type plays a crucial role in N_2O emissions in mountain areas. Annual N_2O emissions from acidic soil were significantly greater than for neutral and alkaline soils under identical climate and agricultural management practices. However, the effects of soil pH on N_2O emissions were inconsistent between wheat and maize seasons. The different soil properties, such as soil pH, availability of C and N substrates, and bacterial ammonia monooxygenase gene abundance were the main drivers of the differences in N_2O emissions across different soils. Overall, soil type affects not only the magnitude but also the thermal sensitivity of N_2O emissions have to be carefully considered for development of mitigation measure and estimate of regional emissions in mountain areas with inherent spatial heterogeneity of soil type.

CRediT authorship contribution statement

Bowen Zhang: Performed field & lab work, Statistical analyses, and Wrote original draft. **Qianying Xiao**: Measured plant N content. **Minghua Zhou**: Funding acquisition, Conceptualization, Supervision and Review & Editing. **Bo Zhu, Tao Wang, Jialiang Tang, Ralf Kiese, Klaus Butterbach-Bahl and Nicolas Brüggemann**: Established the plots and Review & Editing.

Declaration of competing interest

Herewith we confirm that there are no any interest conflicts in the submitted manuscript.

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Appendix A. Supplementary data

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