# **Online Supplemental Material**

Non-cropping period accounting for over a half of annual nitric oxide releases from cultivated calcareous-soil alpine ecosystems with marginally low emission factors

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Supplementary Text S1, Tables S1–S5, and Figure S1

## **Text S1 Materials and methods**

A. Description of experimental site and layout of field treatments

The selected field site (37°36′45″N, 101°18′48″N, 3203 m above sea level) was located aside the Haibei Alpine Meadow Ecosystem Research Station (HAMERS), which is situated in Qinghai Province, China. The site, with a calcareous Matti-Cryic Cambisol soil (Cao et al. 2008), exposures to a continental plateau climate in the temperate zone. Table S1 lists annual mean air temperature and soil properties observed in this study, with the methods for measurement to be given below.

The experimental land (with an area of 0.93 ha) was chosen in a flat landform, thus ensuring relatively uniform soil properties (Table S1). It had been a piece of typical natural alpine *Kobresia humilis* meadow until 1998 and has since been consecutively cultivated with feed oats. The land had been amended with both synthetic fertilizers and yak manure during the first 10 years since its conversion from a meadow to a cultivated land but with only synthetic fertilizers thereafter. During this experimental campaign (from 22 April 2013 to 21 April 2014), the regional typical practices as follows for cultivation of feed oats were adopted. The soil was plowed to a 20 cm depth on 1 June. The oat seeds were sowed at 410 kg ha<sup>-1</sup> (dry seed matter with 8 kg N ha<sup>-1</sup>) on 2 June. Herbicide (2,4-D butyl ester) was sprayed at 488 g ha<sup>-1</sup> prior to oat germination. The aboveground biomass was harvested on 16 September and then air-dried and stored for feeding during winter. There was no irrigation. Except for the four randomly selected field plots for the unfertilized treatment (UF), the remaindering area of the select land was regarded as the fertilized treatment (F) and amended with basal and top-dressing fertilizers. Diammonium hydrogen phosphate and urea, equal to 41 kg N ha<sup>-1</sup> and 20 kg

P ha<sup>-1</sup>, were basally applied simultaneously with oat sowing. The fertilizers and seeds were mechanically incorporated into and mixed with the topsoil (0–5 cm depth) shortly after broadcasting. Top-dressing urea (28 kg N ha<sup>-1</sup>) was broadcasted on 24 July. The nitrogen inputs, in the form of organic nitrogen in the oat seeds and synthetic compounds, totaled 77 kg N ha<sup>-1</sup> yr<sup>-1</sup> for the F treatment. The UF treatment was amended with nitrogen only in form of organic nitrogen in the oat seeds (8 kg N ha<sup>-1</sup>).

Four days before the campaign period, four replicated field plots (each with a size of  $5\times5 \text{ m}^2$  for the F and  $10\times10 \text{ m}^2$  for the UF) for each treatment were randomly established within the select land. Each plot was situated with at least a 2-m distance between the closest boundaries of a plot and the select land and at least a 5-m distance between the closest boundaries of any two plots of the two treatments. Such layouts of the field plots were chosen to minimize the effects of plot/land boundaries and spatial variabilities in soil properties, vegetation and anthropogenic activities, thus facilitating statistical comparisons of the experimental results between the two treatments. The exact location and area of each field plot were fixed during the entire campaign period. As required for field experiments to determine a direct nitrous oxide or nitric oxide (NO) emission factor (IPCC 2006), the UF plots had previously received the same fertilization practices as the F areas but were free from amendment of the synthetic fertilizers only in the year-round experimental period.

Four days before the first observation of NO fluxes, one chamber base frame was installed in the center of each replicated field plot. The base frame remained permanently in each field plot, except for temporary removal and reinstall to allow for soil plowing and follow-up mechanical operations. Each frame was made of stainless steel (each 50 cm long, 50 cm wide and 15 cm high; each wall was 3.0 mm thick) and was inserted fully into the soil; only the upper-edge collar extended out of the soil

surface. A rubber band (6 mm thick) was applied to the upper-edge collar of each base frame for gas-tight sealing of the joint with the chamber. Each chamber was made of stainless steel (50 cm long, 50 cm wide and 40 cm high; each wall was 1.0 mm thick; no bottom). The walls were coated with polystyrene foam boards that were covered with tinfoil to minimize the temperature change within the headspace enclosure during gas sampling. When the vegetation was taller than 40 cm, an alternative chamber with an 80 cm height was adopted to avoid physical damage to the plants within each base frame. There was a tube (7.4 mm inner diameter and 12 cm length) on the top wall of each chamber to allow for an air connection between the headspace and the atmosphere to minimize the pressure difference during sampling. To measure the flux from each plot, a chamber was temporarily mounted onto one of the two base frames to establish a 10-min headspace enclosure for gas sampling.

For the convenience of arranging temporarily intensified observations, a freeze-thaw period (FTP) was defined as a period of at least 5 d during which the daily mean air temperature consecutively fell within the range from -10 to 0 °C. Accordingly, the spring FTP occurred during the period from 21 February to 21 April, 2014 (Fig. S1). The period from the sowing date to the harvest date, the total remaindering time of the full campaign duration, and the full year-round campaign period are referred to as the cropping period, the non-cropping period, and the annual period, respectively.

## **B.** Measurement of nitric oxide fluxes

Nitric oxide fluxes from each field plot were measured during the entire campaign using a technique of combining the chemiluminescence analysis of NO concentrations with gas sampling by opaque, static chambers (Mei et al. 2009; Liu et al. 2009, 2015; Zhang et al. 2018). The flux measurements for all field plots were manually performed daily or every other day during the spring FTP and the periods of 10–14 d following

plowing and fertilizing; otherwise, the measurements were performed once every 3–4 d.

The methods of gas sampling, instrument calibration, and analyses of both NO and nitrogen dioxide (NO<sub>2</sub>) concentrations in the gas samples as well as the flux calculations were in accordance with those detailed by Zhang et al. (2018). The sample NO that was collected into a gas bag until analysis could be converted partly into NO<sub>2</sub> in the presence of ozone. In this regard, the sum of the simultaneously measured NO and NO<sub>2</sub> fluxes was regarded as the measured NO flux. A single NO flux measured by gas sampling during the local time 08:00–10:00 a.m. was used to represent the daily average value (e.g., Liu et al. 2010). According to the instrument precision for NO or NO<sub>2</sub> analysis (0.3 nmol mol<sup>-1</sup> for both gases) and the enclosure time (10 min), the detection limits of NO fluxes for the adopted chamber heights of 40 and 80 cm were 0.4 and 0.8  $\mu$ g N m<sup>-2</sup> h<sup>-1</sup>, respectively.

It should be noted that the NO fluxes measured in this study represented only conservative magnitudes for the investigated ecosystems. The reason is the applied method with a linear-change assumption regarding the gas concentrations in individual static, opaque chamber enclosures might significantly underestimate NO fluxes, e.g., by approximately 31% (ranging 3%–59% at the 95% confidence interval), in comparison with a nonlinear approach (Mei et al. 2009; Yao et al. 2015). The underestimations due to a linear-change assumption indeed do not allow accurate quantification of area-scaled or yield-scaled NO emissions. Nevertheless, the high sensitivity of the applied method to measure NO fluxes allowed investigations of the differences between field treatments and thus determination of direct NO emission factors of applied fertilizer nitrogen (Yao et al. 2015). The high sensitivity also allowed investigation of the regulatory effects of soil variables and other factors on NO fluxes (e.g., Zhang et al. 2018).

C. Auxiliary measurements

The air pressure, air temperature and precipitation were observed and provided by the HAMERS. When the NO fluxes were measured, air temperature within chamber headspace enclosure, topsoil (5 cm depth) temperature ( $T_s$ ) and surface (0–6 cm depth) soil moisture in water-filled pore space (WFPS) were simultaneously measured. The air pressure and headspace air temperature were observed since both variables are involved in the flux calculation to correct the NO gas density. The concentrations of soil (0–10 cm depth) ammonium (NH<sub>4</sub><sup>+</sup>), nitrate (NO<sub>3</sub><sup>-</sup>) and water-extractable organic carbon (WEOC) were observed weekly on one of the days when the NO fluxes were measured. Select soil properties of the (0–20 cm depth), including clay (< 0.002 mm), silt (0.002–0.05 mm), and sand (0.05–2 mm) fractions, soil organic carbon (SOC) and total nitrogen (TN) contents, and soil pH, were measured once in mid-autumn. The mean values were presented for the surface soil (0–6 cm depth) bulk density (BD) that was seasonally measured. The aboveground biomass, which was regarded to approximate the aboveground net primary productivity, and its nitrogen content were also measured at harvest.

To measure the aboveground biomass and its nitrogen content, the harvested plant materials were oven-dried for 30 min at 105  $^{\circ}$ C and then for 48 h at 60  $^{\circ}$ C to obtain the dry matter weight. The nitrogen contents in the dried plant samples were analyzed using the Kjeldahl method (Bao 2000).

The SOC and TN concentrations were analyzed using the potassium dichromate oxidation and the Kjeldahl methods, respectively (Bao 2000). A water-to-soil ratio of 2.5 was used to determine the pH values. The soil particle fractions of different size ranges were measured using Malvern laser particle analysis (Yang et al. 2009).

The headspace air temperature and  $T_s$  were manually measured during gas sampling using digital thermal couples.

The topsoil (0–6 cm) volumetric water content ( $\theta_v$ , cm<sup>3</sup> cm<sup>-3</sup>) was manually measured during the unfrozen periods using a portable frequency-domain reflector moisture meter. During the frozen periods or FTPs, the gravimetric water contents ( $\theta_w$ , g g<sup>-1</sup>) in both ice and liquid phases were measured by oven-drying the soil sample and then the values were converted to  $\theta_v$  units by multiplying with BD (g cm<sup>-3</sup>). Finally, each soil moisture content in WFPS was calculated as WFPS = 100 $\theta_v/(1-BD/2.65)$ .

At each time, four samples were collected (each was a mixture of soil samples from five random points within the corresponding field plot), well mixed, sieved with a 2-mm mesh, and ultimately subsampled via three replicates. On the same day, the subsamples were extracted for analysis of  $NH_4^+$ ,  $NO_3^-$  (1 M potassium chloride, solution-to-soil ratio = 5; shaking for 1 h, and filtering by filter paper) and WEOC (distilled water, water-to-soil ratio = 5; shaking for 1 h, centrifuging for 10 min at 6000 rpm, and filtering by polyethersulfone membrane with < 0.45µm pores) concentrations. Each extract was saved in a 50-mL polyethylene-terephthalate bottle at approximately –18 °C for later assay. The concentrations of  $NH_4^+$ ,  $NO_3^-$  and WEOC in the extracts were analyzed shortly after thawing for 24 h at 4 °C using a continuous flow analyzer (San<sup>++</sup>, Skalar Analytical B.V., The Netherlands).

#### **D.** Data analysis and statistics

The bivariate correlation analysis method was adopted to test the correlations of the NO fluxes during different periods against each of the simultaneously observed soil variables ( $T_s$ , WFPS, and the concentrations of NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and WEOC) and the correlations among these soil variables. For this purpose, the data of NO fluxes and those of each soil variable within a specific period were normalized by their variances using Eq. 1, wherein  $z_{ki}$  and  $x_{ki}$  denote the *i*th values of the *k*th soil variable or NO flux

before and after normalization, respectively, and  $\overline{x_k}$  and  $\sigma_k$  are the mean and the standard variance of the *k*th measurement array. The correlation coefficients between all variable pairs were then calculated, and their significances were tested. These steps were performed by the SPSS 19.0 software package (SPSS Inc., Chicago, USA). The results are reported in a triangle matrix.

$$z_{ki} = (x_{ki} - \overline{x_k}) / \sigma_k \tag{1}$$

Univariate linear and nonlinear regressions were used to investigate the dependences of NO fluxes on the soil variables, using the regression analysis of the SPSS 19.0 software package.

The annual  $EF_d$  (%) and its standard error (SE\_EF<sub>d</sub>) was calculated using Eq. 2 and 3, respectively. In Eqs. 2–3,  $F_N$ ,  $E_F$ ,  $E_{UF}$ , SE<sub>F</sub> and SE<sub>UF</sub> denotes the annual addition rate of synthetic fertilizer nitrogen (kg N ha<sup>-1</sup> yr<sup>-1</sup>), the means of the measured annual NO emissions from the F and UF field plots, and the standard errors for the annual emissions (kg N ha<sup>-1</sup> yr<sup>-1</sup>), respectively.

$$EF_{d} = \frac{E_{F} - E_{UF}}{F_{N}} \times 100$$
(2)

$$SE_{EF_{d}} = 100/F_{N}\sqrt{SE_{F}^{2} + SE_{UF}^{2}}$$
 (3)

In addition to the SPSS 19.0 software package used for statistical analysis, the Origin 8.0 software package (OriginLab Ltd., Guangzhou, China) was used for plotting the data, and the raw experimental data were organized and calculated using the Excel software package of the Microsoft Office Standard 2010 (© 2010 Microsoft Corporation).

The standard errors of means for three to five spatial replicates were given to report the results if not otherwise specified.

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Common information		Both F and	UF		
Latitude		37°36′45″N			
Longitude		101°18′48″E			
Altitude (m)		3203			
Annual mean of air temperature (°C)		-0.1			
Annual total precipitation (mm)		486			
Soil type		Calcareous M	lattic-Cryic		
		Cambisol			
Texture <sup>a</sup>		Silty clay loa	m		
Plow tillage	Date	1 June			
	Depth (cm)	0–20			
Oat sowing	Date	2 June			
	Dry rate (kg $ha^{-1}$ )	410			
	Depth (cm)	0–5			
Herbicide spray	Туре	2,4-D butyl e	ster		
	Date	25 June			
	Rate (kg N ha <sup>-1</sup> )	0.488			
Harvest	Date	26 Septembe	r		
	Method	Manual operation	ation		
Residue retention as stubbles	Fraction	0.05			
Treatment-specific information		F	UF		
Sand (0.05–2 mm) fraction (%) $^{a,b,c}$		32 (1)	36(1)		

**Table S1** Select soil properties and other natural or management information of theexperimental site and the fertilized (F) and unfertilized (UF) field treatments.

Silt (0.002–0.05 mm) fraction (%) <sup>a.</sup>	,b,c	51 (1)	49(1)
Clay (< 0.002 mm) fraction (%) $^{a,b,c}$		17 (1)	15(1)
Organic carbon content (g C kg <sup><math>-1</math></sup> d.s.) <sup>a,b,</sup>	,c	46 (2)	46 (2)
Total nitrogen content (g N kg <sup><math>-1</math></sup> d.s.) <sup>a,b,c</sup>		4.4 (0.1)	5.1 (0.4)
pH (H <sub>2</sub> O) <sup>a,b,c</sup>		8.3 (0.3)	8.3 (0.2)
Bulk density (g cm <sup>-3</sup> ) <sup>a,b,d</sup>		0.88 (0.04)	0.88 (0.04)
Aboveground net primary productivity (§	$g m^{-2})^{a,b}$	1068 (38)	901 (120)
Plant nitrogen content $(g kg^{-1})^{a,b}$		8.6 (0.3)	9.0 (0.3)
Aboveground nitrogen uptake (kg N ha <sup>-1</sup>	91.8 (3.1)	81.1 (10.8)	
Basal urea	Date	2 June	2 June
	Depth(cm)	0–5	0–5
	Rate (kg N ha <sup>-1</sup> )	23	0
Basal diammonium hydrogen phosphate	Date	2 June	2 June
	Depth(cm)	0–5	0–5
	Rate (kg N ha <sup>-1</sup> )	18	0
	Rate (kg P ha <sup><math>-1</math></sup> )	20	0
Basal organic nitrogen in seeds	Date	2 June	2 June
	Depth(cm)	0–5	0–5
	Rate (kg N ha <sup>-1</sup> )	8	8
Top-dressed urea	Date	24 July	24 July
	Depth(cm)	0	0
	Rate (kg N ha <sup>-1</sup> )	28	0

<sup>a</sup> Data observed in this study. <sup>b</sup> Given data are means (standard errors) of 3–5 spatial replicates. <sup>c</sup> and <sup>d</sup>, for 0–20 and 0–6 cm soil depths, respectively.

**Table S2** Precipitation (*P*), soil (5 cm) temperature ( $T_s$ ), soil (0–6 cm) moisture in water-filled pore space (WFPS), and concentrations of soil (0–10 cm depth) ammonium ([NH<sub>4</sub><sup>+</sup>]), nitrate ([NO<sub>3</sub><sup>-</sup>]) and water-extractable organic carbon ([WEOC]) during the freeze–thaw period (FTP), non-cropping period (NCP), cropping period (CP), and annual period (AP).

	<i>P</i> (mm)			WFPS (%)	WFPS (%)		$[NH_4^+](mg N kg^{-1} d.s.)$		$[NO_3^{-}](mg N kg^{-1} d.s.)$		$[WEOC] (mg C kg^{-1} d.s.)$	
		F	UF	F	UF	F	UF	F	UF	F	UF	
FTP	76	-0.7 (1.4)	0.0 (1.5)	30.9 (4.3)	31.7 (4.7)	1.4 (0.5)	2.2 (1.0)	20.5 (2.7)	26.0 (5.6)	73.9 (4.6)	97.0 (12.0)	
NCP	166	$-3.3(1.2)^{\text{B}}$	$-2.5(1.1)^{\text{B}}$	31.3 (2.6) <sup>B</sup>	31.6 (2.0) <sup>B</sup>	$0.7 (0.5)^{B}$	1.2 (0.7)	18.1 (1.5)	18.5 (2.2) <sup>B</sup>	75.0 (4.9)	83.0 (4.6) <sup>B</sup>	
СР	320	10.8 (0.7) <sup>A</sup>	11.0 (0.7) <sup>A</sup>	40.7 (2.2) <sup>A</sup>	39.3 (2.2) <sup>A</sup>	4.1 (1.8) <sup>A</sup>	1.6 (0.4)	12.5 (4.6)	8.9 (3.0) <sup>A</sup>	95.5 (8.0)	104.7 (9.0) <sup>A</sup>	
AP	486	3.3 (1.1)	3.7 (1.1)	35.6 (1.6)	35.2 (1.5)	1.8 (0.7)	1.3 (0.5)	16.2 (1.8)	15.2 (1.8)	81.4 (3.6)	89.8 (4.4)	

For the full treatment names, refer to Table S1. FPT and CP were from 21 February to 21 April, and 2 June to 26 September, 2014, respectively. The given data are means of 3–5 spatial replicates, with standard errors showing within the parentheses. The different superscript capital letters indicate the significant differences between CP and NCP within a treatment at p < 0.05, respectively.

	Fertiliz	ed treatm	ent (F)			Unfertilized treatment (UF)				Both F and UF					
	NO	T <sub>s</sub>	WFPS	[NH4 <sup>+</sup> ]	[NO <sub>3</sub> <sup>-</sup> ]	NO	T <sub>s</sub>	WFPS	[NH4 <sup>+</sup> ]	[NO <sub>3</sub> <sup>-</sup> ]	NO	T <sub>s</sub>	WFPS	[NH4 <sup>+</sup> ]	[NO <sub>3</sub> <sup>-</sup> ]
NCP															
T <sub>s</sub>	0.70***					0.66**					0.68**				
WFPS	0.10	0.14				0.16	0.16				0.13	0.15			
$[\mathrm{NH_4}^+]$	-0.06	-0.02	-0.02			0.03	-0.10	-0.04			0.01	-0.06	-0.03		
[NO <sub>3</sub> <sup>-</sup> ]	0.30	0.25	0.35*	-0.17		0.33	0.18	0.33	0.35*		0.31**	0.20	0.33**	0.22	
[WEOC]	0.19	0.46**	-0.01	-0.08	0.14	0.22	0.30	0.02	0.28	0.50**	0.21	0.37**	0.01	0.19	0.38**
СР															
T <sub>s</sub>	0.36*					<b>0.45</b> *					<b>0.41</b> <sup>**</sup>				
WFPS	-0.34	-0.23				-0.30	-0.16				-0.31*	-0.19			
$[\mathrm{NH_4}^+]$	0.41	-0.12	-0.36			0.30	-0.27	-0.25			0.32	-0.12	-0.25		
[NO <sub>3</sub> <sup>-</sup> ]	<b>0.48</b> <sup>*</sup>	0.06	-0.47*	0.92**		0.37	0.06	-0.49*	0.79**		0.42*	0.05	-0.45**	0.84**	

Table S3 Coefficient matrix of the correlations among nitric oxide (NO) fluxes and the simultaneously observed soil variables.

[WEOC] 0.08	0.52*	-0.17	-0.16	0.03	0.13	0.47	-0.37	-0.19	0.13	0.10	0.49**	-0.28	-0.16	0.05
AP														
$T_{\rm s}$ <b>0.42</b> <sup>*</sup>	k				0.39**					0.40**				
WFPS -0.10	0.28*				-0.09	0.26*				-0.10	0.27**			
[NH <sub>4</sub> <sup>+</sup> ] <b>0.45</b> <sup>*</sup>	* 0.17	-0.11			0.15	0.09	-0.10			0.32**	0.13	-0.10		
[NO <sub>3</sub> <sup>-</sup> ] <b>0.33</b> *	-0.07	-0.16	0.62**		0.17	-0.18	-0.04	0.33*		0.25*	-0.12	-0.10	0.49**	
[WEOC] 0.18	0.53**	0.05	-0.04	-0.06	0.19	0.43**	-0.04	0.21	0.20	0.18	0.48**	0	0.07	0.09

For the details on  $T_s$ , WFPS, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and WEOC, refer to Table S2. \* and \*\* indicate the significances at p < 0.05 and 0.01, respectively.

No.	Equation	$r^2$	п	р	$Q_{10}$	Remarks
1	$F_{\rm NO} = 1.99e^{0.162T_{\rm s}}$	0.67	34	< 0.01	5.1	F: NCP
2	$F_{\rm NO} = 0.84 e^{0.133 T_{\rm s}}$	0.27	31	< 0.01	3.7	F: CP
3	$F_{\rm NO} = 0.080[{\rm NO_3}^-]e^{0.130T_{\rm s}}$	0.52	18	< 0.01	3.7	F: CP
4	$F_{\rm NO} = 0.04 [{\rm NH_4}^+] e^{0.318T_{\rm s}}$	0.54	17	< 0.01	24.0	F: CP
5	$F_{\rm NO} = (0.22[\rm NH_4^+] + 0.02[\rm NO_3^-] - 0.01[\rm WEOC] - 0.08\rm WFPS + 4.30) e^{0.133T_s}$	0.75	15	< 0.01	3.7	F: CP
6	$F_{\rm NO} = 1.22e^{0.117T_{\rm s}}$	0.31	65	< 0.01	3.3	F: AP
7	$F_{\rm NO} = 0.082[{\rm NO_3}^-]e^{0.129T_{\rm s}}$	0.56	52	< 0.01	3.6	F: AP
8	$F_{\rm NO} = (0.167[\rm NH_4^+] + 0.036[\rm NO_3^-] - 0.013[\rm WEOC] - 0.061\rm WFPS + 4.14)e^{0.134T_{\rm s}}$	0.76	49	< 0.01	3.8	F: AP
9	$F_{\rm NO} = 2.06e^{0.156T_{\rm s}}$	0.55	34	< 0.01	4.8	UF: NCP
10	$F_{\rm NO} = 0.39 e^{0.172T_{\rm s}}$	0.45	31	< 0.01	5.5	UF: CP
11	$F_{\rm NO} = 0.08[{\rm NO_3}^-]e^{0.144T_{\rm s}}$	0.62	18	< 0.01	4.2	UF: CP
12	$F_{\rm NO} = 0.17 [{\rm NH_4}^+] e^{0.282T_{\rm s}}$	0.64	18	< 0.01	16.8	UF: CP

**Table S4** Relationships between nitric oxide (NO) fluxes ( $F_{NO}$ ,  $\mu g N m^{-2} h^{-1}$ ) and simultaneously observed soil variables.

13	$F_{\rm NO} = (0.48[{\rm NH_4^+}] + 0.03[{\rm NO_3^-}] - 0.002[{\rm WEOC}] - 0.03{\rm WFPS} + 1.32)e^{0.177T_{\rm s}}$	0.75	16	< 0.01	5.9	UF: CP
14	$F_{\rm NO} = 0.65 e^{0.151 T_{\rm s}}$	0.42	65	< 0.01	4.5	UF: AP
15	$F_{\rm NO} = 0.08[{\rm NO_3}^-]e^{0.143T_{\rm s}}$	0.62	52	< 0.01	4.2	UF: AP
16	$F_{\rm NO} = (0.37[\rm NH_4^+] + 0.05[\rm NO_3^-] - 0.01[\rm WEOC] - 0.04\rm WFPS + 2.41) e^{0.164T_{\rm S}}$	0.74	50	< 0.01	5.1	UF: AP
17	$F_{\rm NO} = 2.02 e^{0.159T_{\rm s}}$	0.60	68	< 0.01	5.0	F+UF: NCP
18	$F_{\rm NO} = 0.56e^{0.154T_{\rm s}}$	0.36	62	< 0.01	4.5	F+UF: CP
19	$F_{\rm NO} = (0.20 [\rm NH_4^+] + 0.06[\rm NO_3^-] - 0.003[\rm WEOC]) e^{0.133T_s}$	0.67	31	< 0.01	3.8	F+UF: CP
20	$F_{\rm NO} = 0.89 e^{0.134 T_{\rm s}}$	0.37	130	< 0.01	3.8	F+UF: AP
21	$F_{\rm NO} = (0.19[{\rm NH_4^+}] + 0.07[{\rm NO_3^-}] - 0.003[{\rm WEOC}])e^{0.126T_{\rm s}}$	0.67	99	< 0.01	3.5	F+UF: AP
22	$F_{\rm NO} = (0.17[\rm NH_4^+] + 0.05[\rm NO_3^-] - 0.01[\rm WEOC] - 0.06\rm WFPS + 3.78) e^{0.135T_s}$	0.72	99	< 0.01	3.9	F+UF: AP

F, UF and F+UF represent the fertilized, unfertilized and both treatments, respectively. For the details on the different periods and soil variables, refer to Table S2.  $r^2$ , n, p and  $Q_{10}$  denote the coefficient of determination, sample size, significance level, and temperature sensitivity coefficient, respectively. A given  $Q_{10}$  value represents the fold of the changes in NO fluxes due to a 10-degree change in soil temperature.

**Table S5** Statistics of the annual nitric oxide (NO) emissions from fertilized (A) and short-term unfertilized (B) arable uplands and annual direct NO emission factors (C) reported in the literature

A. Annual NC	emissions	from	fertilized	arable uplands.
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Conditions	NO emission	Remarks		
	Range	Mean	SD	_
Worldwide, non-alpine	0.2–23	1.1	-	[1-2]
Non-alpine, calcareous soils, China	0.7–5.7	1.7	1.2	[3–10]
Non-alpine, non-calcareous soils,	6.6–47.1	15.2	12.6	[11–16]
vegetable fields, tea gardens, China				

**B.** Annual NO emissions from short-term unfertilized (usually not fertilized only in the current year) arable uplands.

Conditions	NO emission	Remarks		
	Range	Mean	SD	_
Non-alpine, calcareous soils, China	0.26-0.60	0.45	0.14	[3–10]
Non-alpine, non-calcareous soils,	0.33–2.80	0.73	0.58	[11–16]
vegetable fields, tea gardens, China				

**C.** Direct NO emission factors  $(EF_d)$  of arable uplands.

Conditions	EF <sub>d</sub> (%)	$\mathrm{EF}_{\mathrm{d}}$ (%)					
Conditions	Range	Mean	SD	_ Remarks			
Cultivated grasslands, Japan	0.01-0.36	-	-	[17]			
Cultivated savannas	0.60-1.5	-	-	[18]			

Global fertilized lands	0.5–0.7	-	-	[19–20]
National fertilized lands, China	~ 0.67	-	-	[12]
Non-alpine, calcareous soils, China	0.08-0.78	0.30	0.16	[3–10]
Non-alpine, non-calcareous soils,	2.2–4.0	3.2	0.6	[11–16]
vegetable fields, tea gardens, China				

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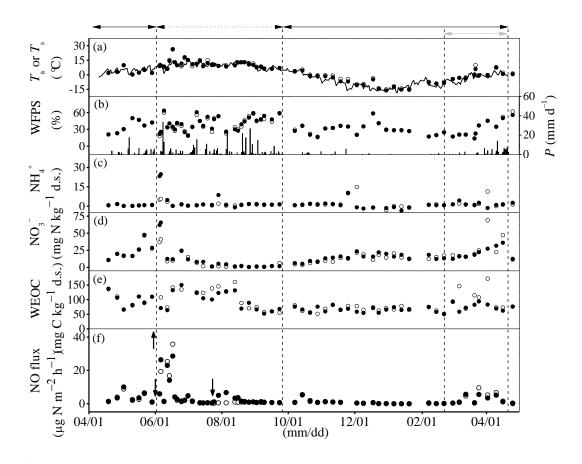
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**Fig. S1** Year-round dynamics of precipitation, air temperature, select soil variables, and nitric oxide (NO) fluxes. The horizontal arrows with grey solid, black solid and dashed lines indicate the freeze–thaw period, non-cropping period and cropping period, respectively. For the details on the soil variables, refer to Table S2. The solid and empty dots are for the fertilized and unfertilized treatments, respectively. a. Temperature of air ( $T_a$ , line) and soil ( $T_s$ ). b. WFPS and precipitation (P, bar). c–e. NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and WEOC concentrations, respectively. f. NO fluxes. The standard errors of four spatial replicates are not shown for clarity. The upward and downward arrows in the bottom panel indicate the plow tillage and fertilization dates of the fertilized treatment, respectively.