

Original Article

Simultaneous quantification of greenhouse gas and nitric oxide emissions from subtropical conventional vegetable systems: a 2-site field case study in Sichuan Basin

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Abstract: Quantification of greenhouse gases [nitrous oxide (N₂O) and methane (CH₄)] and nitric oxide (NO) emissions from subtropical conventional vegetable systems through multi-site field measurements are needed to obtain accurate regional and global estimates. N₂O, NO and CH₄ emissions from subtropical conventional vegetable systems were simultaneously measured at two different sites with hilly topography in the Sichuan basin, southwest China by using the static chamber gas chromatography technique. Results showed that annual soil N₂O and NO fluxes for the treatment receiving N fertilizer ranged from 6.34-7.71 kg N ha⁻¹ yr⁻¹ and 0.69-0.85 kg N ha⁻¹ yr⁻¹, respectively, while decreased soil CH₄ uptakes by 26.4% as compared with no N fertilizer addition across our two sites of experiment. Overall, the average direct N₂O and NO emission factor (EF_d) were 0.71% and 0.12%, respectively, which were both lower than the available EF_d for subtropical conventional vegetable systems. This finding indicates that current regional and global

estimates of N₂O and NO emissions from vegetable fields are likely overestimated. Background N₂O emissions (3.42-3.62 kg N ha⁻¹ yr⁻¹) from the subtropical conventional vegetable systems were relatively high as compared with available field measurements worldwide, suggesting that background N₂O emissions cannot be ignored for regional estimate of N₂O emissions in subtropical region. Nevertheless, the significantly intra- and inter-annual variations in N₂O, CH₄ and NO emissions were also observed in the present study, which could be explained by temporal variations of environmental variables (i.e. soil temperature and moisture). The differences in N₂O and NO EF_d and CH₄ emissions between various vegetable systems in particular under subtropical conditions should be taken into account when compiling regional or global inventories and proposing mitigation practices.

Keywords: Subtropical vegetable system; Nitrous oxide; Nitric oxide; Methane; Emission factor; Background emissions

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1 Introduction

The increases in atmospheric nitrous oxide (N₂O), nitric oxide (NO) and methane (CH₄) concentrations contribute greatly to the global climate change and atmospheric environmental deterioration (IPCC 2014). Agricultural production activities have largely contributed to the increases in atmospheric N₂O and NO concentrations (Bouwman et al. 2002b; Liu et al. 2017). In particular, nitrogen (N) fertilizer application has been identified as the dominant contributor of N₂O and NO emissions from agriculture soils, being responsible for 60% and 10% of the total global anthropogenic emissions, respectively (IPCC 2014). Globally, the atmospheric CH₄ sinks of aerobic soils have been estimated to an average of -28.0 Tg CH₄ yr⁻¹ and the subtropical region accounted for 43.2% (Cai 2012). Nevertheless, the uncertainties in regional and global estimates of N₂O, NO and CH₄ emissions are still high, most likely due to the great spatial-temporal variability among different soil type, cropping system, climate and management practice (Bouwman et al. 2002a; Butterbach-Bahl et al. 2013; Jia et al. 2012; Rashti et al. 2015). Therefore, multi-site field measurements of soil N₂O, NO and CH₄ fluxes under different agricultural practices are needed, in particular for intensively used agricultural regions under subtropical climate.

China is the largest vegetable cultivation country worldwide, accounting for 52.0% of the global vegetable cultivation areas (FAO 2017). In China, most vegetable cultivation systems are intensively managed with high N fertilizer inputs [may up to 1568 kg N ha⁻¹ yr⁻¹ (Mei et al. 2011)]. Thus, Chinese vegetable cultivation systems were generally identified as the “hot spots” of soil N₂O and NO emissions (Pang et al. 2009; Rashti et al. 2015; Wang et al. 2018a; Yao et al. 2015), and might have great GHG mitigation potential. However, there are few studies on monitoring of soil N₂O and/or NO emissions from subtropical vegetable cultivation systems relative to cereal cropping systems (e.g., Della Chiesa et al. 2019; Liu et al. 2017). Furthermore, the gaps in datasets on simultaneous measurement of soil N₂O and NO emissions from vegetable cultivation systems somehow constrain the accuracy of regional and global estimates of soil N₂O and NO emissions.

The aerobic soils are important sinks for atmospheric CH₄ (Cai 2012; Dutaur & Verchot 2007). Although the applied fertilizer N may mediate the

microbial activities of methanogens and methanotrophs for CH₄ production and consumption processes in agricultural soils, the direction and magnitude of N fertilization effects on soil CH₄ fluxes remain uncertain (Gulledge et al. 2004; Sitaula et al. 2000; Tate 2015; Zhou et al. 2014). For example, some studies reported that N fertilization inhibited soil CH₄ oxidation capacity, because of the competitive inhibition of ammonium on methane monooxygenase, most likely due to the toxicity/osmotic effect of hydroxylamine and nitrite on methanotrophic bacteria (Sitaula et al. 2000). On the other hand, some studies showed that N fertilization enhanced soil CH₄ oxidation because nitrifiers can stimulate CH₄ oxidation and alleviate of N limitation for methanotrophs in the N limitation ecosystems (Chan & Parkin 2001; Hellebrand et al. 2003). Nevertheless, the N fertilization effects on soil CH₄ fluxes have not been well documented in the vegetable cultivation systems.

Therefore, we complied field experiments at two different study sites to simultaneously measure soil N₂O, NO and CH₄ fluxes as well as environmental variables from the subtropical conventional vegetable cultivation systems. The objectives of the present study were to fill gaps in datasets of multi-site field simultaneous measurements of N₂O, NO and CH₄ fluxes, (2) to obtain direct emission factors (EF_i) and background emissions of soil N₂O and NO, and (3) to quantify N fertilization effects on soil CH₄ fluxes from the subtropical conventional vegetable cultivation systems.

2 Materials and Methods

2.1 Field site description and experimental design

The field study was conducted at two subtropical conventional vegetable fields (over 20 years consecutive vegetable cultivation history before our experiment), separately located at Wenjiang in the suburban area of Chengdu (CD, 30°41' N, 103°48' E) with plain topography and the Yanting Agro-Ecological Station of Chinese Academy of Sciences (YT, 31°16' N, 105°28' E) with hilly topography in the Sichuan basin, southwest China. The climate for the CD and YT sites is classified as moderate subtropical monsoon climate. The mean annual precipitation over

the past 30 years were 868 mm for the CD site and 863 mm for the YT site. The mean annual air temperature were 16.0°C for the CD site and 17.3°C for the YT site. The chemical-physical properties of topsoil (0-20 cm) for the CD and YT sites were shown in Table 1.

Table 1 The properties of topsoil (0-20 cm) at the Chengdu and Yanting experimental sites.

Properties	Experimental site	
	Chengdu	Yanting
Soil type	Cambisols	Eutric Regosols
pH (H ₂ O)	7.37	8.35
Soil organic matter (g kg ⁻¹)	30.18	27.50
Total nitrogen (g kg ⁻¹)	1.87	1.34
Olsen phosphorus (mg kg ⁻¹)	22.65	130.30
Available potassium (mg kg ⁻¹)	56.42	460.90
Bulk density (g cm ⁻³)	1.23	1.21
Clay (<0.002 mm) %	8.70	8.30
Silt (0.002-0.05 mm) %	43.70	43.10
Sand (>0.05 mm) %	47.60	48.40

The field experiments were performed from November 2004 to October 2006 at CD and from October 2006 to May 2008 at YT. Two treatments (non-N fertilizer and N fertilized) were arranged with three replicates based on a randomized block design at each site. The plot size was 15 m² (3 m × 5 m) for both experimental sites. The details of vegetable cultivation and agricultural management practices for the two experimental sites are shown in Table 2. In accordance with the locally conventional cultivation practices, the N, P and K fertilizers were broadcasted as basal fertilizer followed by plowing with the topsoil before vegetable crops were transplanted or sown. Four different vegetable species were successively grown at the two sites, including Baozijing [*Brassica juncea*. var. *Gemmifera* L. (BJGL)], lettuce (*Lactuca sativa* L.), Chinese cabbage (*Beassica pekinensis*Rupr.) and cabbage (*Brassica deracea* var. *capitate* L.).

2.2 Measurements of soil N₂O, NO and CH₄ fluxes

The soil N₂O, NO and CH₄ emissions from the vegetable fields of CD and YT were simultaneously monitored by using the static chamber-gas chromatography technique/chemiluminescent analyzer method (Zheng et al. 2008; Zhou & Butterbach-Bahl 2014). Stainless steel square chamber bases (80 cm × 80 cm) were pre-installed into the soil to a depth of 15 cm for each plot before

vegetable transplanting/sowing and kept undisturbed over each vegetable season, there is a groove (3 cm in both width and depth) on the top edge of each base used for water seal. Stainless steel chambers (80 cm × 80 cm × 50 cm) were wrapped with thermo-insulated layer to minimize air temperature variations inside the chamber, besides, two fans (10 cm in diameter, continuously working throughout the whole period of gas sampling to mix the headspace air) and one thermometer probe (to measure the air temperature) were also equipped in each chamber. To avoid the disturbance of sampling activities to experiment plots, a wooden walking bridge was set up in each plot before each vegetable season. The gas sampling frequency was every other day for approximately two weeks following N fertilization and then changed to twice per week throughout each vegetable season and fallow season.

The gas sampling time for both sites was 9:00 a.m.-11:00 a.m. (Beijing time) at a 7-min interval (0, 7, 14, 28 min after chamber closure), gas samples were taken from the headspace of each chamber by using 60 mL medical syringes. All gas samples were analyzed within 8 hours by using a gas chromatography (HP 5890II, Hewlett-Packard, Palo Alto, California, USA) equipped with an electron capture detector (ECD) for N₂O detection and a flame ionization detector (FID) for CH₄ detection. The N₂O and CH₄ fluxes were calculated based on the change rate of gas concentrations in the enclosed chamber headspace over time.

To determine the NO fluxes, two gas samples (approximately 5L) were collected at the beginning and end of the same chamber enclosure period that was used for N₂O and CH₄ measurements. Gas samples were collected by using pre-evacuated inert aluminum-coated plastic bags (Guangming Research & Design Institute of Chemical Industry, Dalian, China) and a vacuum pump with a flow rate of 2-3 L min⁻¹. The gas samples were analyzed immediately to determine the NO concentration using a chemiluminescence NO-NO₂-NO_x analyzer (Monitor labs 8840, USA). The NO fluxes were also calculated based on the change rate of NO concentration in the enclosed chamber headspace over time.

The cumulative emissions of N₂O, NO and CH₄ were determined by linear interpolation of the daily fluxes between the two closest days when observations were taken. The direct N₂O and NO emission factors (EF_d, %) were calculated by the equation

Table 2 Vegetable cultivation and fertilizer [N (urea, 46.4% N), P (superphosphate, 12% P₂O₅) and K (potassium chloride, 60% K₂O)] application practices at the Chengdu and Yanting experimental sites throughout the experimental period.

Site	Vegetation	Transplanting /sowing	Harvest	Basal fertilization	N (kg N ha ⁻¹)	P (kg P ₂ O ₅ ha ⁻¹)	K (kg K ₂ O ha ⁻¹)	Topdressing	N (kg N ha ⁻¹)	Row×plant spacing
Cheng du	BJGL ^a	Nov.25 2004	Mar.28 2005	Nov.29 2004	100	200	160	Jan.6 2005	100	30 cm×35 cm
	Lettuce	Apr.10 2005	May.25 2005	Apr.11 2005	100	200	160	May.10 2005	100	30 cm×40 cm
	Fallow period	May.26 2005	Sep.27 2005	-	-	-	-	-	-	-
	BJGL	Sep.28 2005	Feb.13 2006	Oct.19 2005	100	200	160	Dec.15 2005	100	30 cm×35 cm
Yan ting	Cabbage	Feb.19 2006	May.15 2006	Mar.8 2006	100	200	160	Apr.18 2006	100	45 cm×50 cm
	Fallow period	May.16 2006	Oct.25 2006	-	-	-	-	-	-	-
	BJGL ^a	Oct.25 2006	Feb.11 2007	Nov.7 2006	90	150	160	Dec.10 2006	60	30 cm×35cm
	Fallow period	Feb.12 2007	Apr.9 2007	-	-	-	-	-	-	-
Yan ting	Lettuce	Apr.10 2007	Jun.4 2007	Apr.24 2007	90	150	160	May.2 2007	60	35 cm×40 cm
	Chinese cabbage	Jun.5 2007	Aug.22 2007	Jun.5 2007	90	150	160	Jul.10 2007	60	45 cm×50 cm
	Cabbage	Aug.27 2007	Oct.26 2007	Sep.12 2007	90	150	160	Oct.9 2007	60	45 cm×50 cm
	Fallow period	Oct.27 2007	May.6 2008	-	-	-	-	-	-	-

Note: - means no fertilization or plantation were taken during fallow period. ^aBJGL: *Brassica juncea*. var. *Gemmifera L.*

Table 3 Seasonal and annual cumulative nitrous oxide (N₂O), nitric oxide (NO) and methane (CH₄) fluxes (Mean ± SE), direct emission factors (EF_d, in %), and the ratio of NO/N₂O at the Chengdu and Yanting sites throughout the experimental period

Site	Period	N ₂ O (kg N ha ⁻¹)		NO (kg N ha ⁻¹)		CH ₄ (kg C ha ⁻¹)		NO/N ₂ O		EF _d (%)	
		Fertilized	Control	Fertilized	Control	Fertilized	Control	Fertilized	Control	N ₂ O	NO
Cheng du	BJGL	3.66±0.40 a	1.39±0.34 b	0.79±0.33 a	0.02±0.01 b	-1.07±0.11 a	-1.04±0.21 a	0.22	0.01	1.14	0.39
	Lettuce	2.17±0.18 a	1.60±0.24 b	0.16±0.01 a	0.09±0.02 b	-0.61±0.19 a	-0.94±0.30 b	0.07	0.06	0.29	0.04
	Fallow period	1.44±0.32 a	0.68±0.08 b	n.m.	n.m.	-0.71±0.11 a	-0.65±0.36 a	-	-	-	-
	BJGL	2.25±0.17 a	1.67±0.18 b	n.m.	n.m.	-0.71±0.37 a	-1.73±0.40 b	-	-	0.29	-
Yan ting	Cabbage	1.51±0.08 a	0.78±0.23 b	n.m.	n.m.	-0.65±0.31 a	-1.01±0.35 a	-	-	0.37	-
	Fallow period	1.63±0.62 a	0.71±0.11 b	n.m.	n.m.	-0.71±0.08 a	-0.66±0.54 a	-	-	-	-
	Annual	6.34±0.42 a	3.42±0.21 b	-	-	-2.23±0.07 a	-3.02±0.34 b	-	-	0.73	-
	BJGL	1.49±0.23 a	0.57±0.01 b	0.41±0.08 a	0.01±0.01 b	-0.39±0.08 a	-0.34±0.13 a	0.28	0.02	0.61	0.27
Yan ting	Fallow period	0.21±0.01 a	0.22±0.03 a	0.01±0.01 a	0.02±0.00 a	-0.18±0.07 a	-0.17±0.09 a	0.05	0.09	-	-
	Lettuce	1.07±0.24 a	0.69±0.14 b	0.16±0.02 a	0.02±0.01 b	-0.25±0.22 a	-0.53±0.07 b	0.15	0.03	0.25	0.09
	Chinese cabbage	3.59±0.31 a	1.38±0.31 b	0.10±0.02 a	0.02±0.01 b	-0.25±0.09 a	-0.45±0.03 b	0.03	0.01	1.47	0.05
	Cabbage	1.42±0.18 a	0.76±0.11 b	0.09±0.03 a	0.01±0.01 b	-0.13±0.12 a	-0.17±0.06 a	0.06	0.01	0.44	0.05
Annual	Fallow period	2.36±0.40 a	1.24±0.19 b	0.03±0.00 a	0.02±0.01 a	-0.73±0.21 a	-1.32±0.06 b	0.01	0.02	-	-
	Annual	7.71±0.07 a	3.62±0.15 b	0.75±0.07 a	0.08±0.01 b	-1.22±0.29 a	-1.66±0.03 b	0.1	0.02	0.68	0.12

Note: n.m. means no measurements, - means calculation cannot be done. Different lowercase letters indicate significant differences between fertilized and control treatments within each season at p<0.05 level.

($EF_d = (F_N - F_0)/N \times 100\%$). Here, F_N is the cumulative flux of N_2O or NO ($kg\ N\ ha^{-1}$) from the fertilized treatment, F_0 is the cumulative flux of N_2O or NO ($kg\ N\ ha^{-1}$) from the control treatment, and N is the N application rate ($kg\ N\ ha^{-1}$). It should be noted that the annual N_2O , NO and CH_4 fluxes were calculated by the cumulative emissions from Nov. 25 2004 to Nov. 25 2006 for the CD site and from Oct. 25 2006 to Oct. 26 2007 for the YT site, the cumulative emissions from Oct. 25 2006 to Nov. 25 2006 (the last fallow season) were not measured and calculated by linear interpolation method. The N_2O emissions from April.11 2005 to May.8 2006 for the CD site and from Oct.25 2006 to Oct.26 2007 for the YT site were adopted and reprocessed from Yu et al. (2008) and Yu et al. (2012), respectively.

2.3 Auxiliary measurement

Daily precipitation and air temperature were recorded by the established automatic meteorological stations near the experimental plots at the CD and YT sites. The soil temperature (at 5 cm depth) was monitored by using a manual thermometer (JM624 Tianjin Jinming Instrument Co. Ltd) and the soil moisture (0-5 cm) was monitored by using a portable frequency domain reflectometry (FDR) moisture sensor (MPKit-B Hangzhou Tuopu Instrument Co. Ltd) when taking gas samples. The measured soil moisture by FDR was converted to water-filled pore space (WFPS) by the equation ($WFPS = \text{soil volumetric water content} / (1 - \text{soil bulk density} / 2.65) \times 100\%$), here, the assumed soil particle density was $2.65\ g\ cm^{-3}$.

2.4 Data processing and statistical analysis

The statistical data analyses were carried out with SPSS 19.0 software program (SPSS Inc, 2008). Significant differences in the seasonal (each crop or fallow season) and annual N_2O , NO and CH_4 fluxes between the fertilized and control treatments were compared by the one-way ANOVA with Tukey's range test. The relationships between N_2O , NO and CH_4 emissions of per sampling date and environmental factors (soil temperature and WFPS) were analyzed using linear regression. All the plots were prepared using the Origin 9.4 software (Origin Lab Corporation Northampton, USA).

3 Results

3.1 Environmental condition

The annual precipitations were 860.0 mm for the CD site and 877.7 mm for the YT site, and the average daily air temperature were $16.9^\circ C$ and $17.6^\circ C$ for the CD and YT sites, respectively (Fig. 1 a-b). The temporal patterns of soil temperature (at 5 cm depth) and soil moisture (water-filled pore space [WFPS] at 5 cm depth) were comparable with temporal patterns of daily air temperature and precipitation. The average soil temperature and WFPS were $16.1^\circ C$, 59.1% and $16.5^\circ C$, 59.6% for the CD and YT sites, respectively (Fig. 1 c-d and e-f).

3.2 N_2O fluxes

At the CD site, soil N_2O emissions fluctuated from 4.7 to $374.7\ \mu g\ N\ m^{-2}\ h^{-1}$ for the N fertilization treatment and from -55.0 to $346.3\ \mu g\ N\ m^{-2}\ h^{-1}$ for the control (Fig. 2). The pulses of N_2O emission (mean: $231.0\ \mu g\ N\ m^{-2}\ h^{-1}$) were consistently observed within approximately two weeks following N fertilization events. Compared to the control, N fertilizer application significantly increased seasonal and annual cumulative N_2O emissions. On average, annual N_2O fluxes for the N fertilization treatment ($6.34\ kg\ N\ ha^{-1}\ yr^{-1}$) were approximately two times higher than those for the control ($3.42\ kg\ N\ ha^{-1}\ yr^{-1}$) and the fallow period of fertilization treatment accounted for 24.2% of annual N_2O fluxes (Table 3). The seasonal direct N_2O emission factor (EF_d) were in the range of 0.29% to 1.14% (mean: 0.73%).

At the YT site, the N_2O pulses averaged $438.9\ \mu g\ N\ m^{-2}\ h^{-1}$, soil N_2O emissions ranged from 0.6 to $1229.6\ \mu g\ N\ m^{-2}\ h^{-1}$ for the N fertilization treatment and from 0.4 to $402.4\ \mu g\ N\ m^{-2}\ h^{-1}$ for the control (Fig. 3). The seasonal cumulative N_2O fluxes ranged from 0.21 to $3.59\ kg\ N\ ha^{-1}$ for the N fertilization treatment and ranged from 0.22 to $1.38\ kg\ N\ ha^{-1}$ for the control (Table 3). The annual N_2O fluxes were in the range of 7.69-7.90 $kg\ N\ ha^{-1}\ yr^{-1}$ (mean: $7.71\ kg\ N\ ha^{-1}\ yr^{-1}$) for the N fertilization treatment and 3.38-3.90 $kg\ N\ ha^{-1}\ yr^{-1}$ (mean: $3.62\ kg\ N\ ha^{-1}\ yr^{-1}$) for the control. The vegetable growing periods of fertilization treatment on average accounted for 74.7% of the annual cumulative N_2O fluxes. The seasonal EF_d of N_2O ranged from 0.25% to 1.47% (mean: 0.68%).

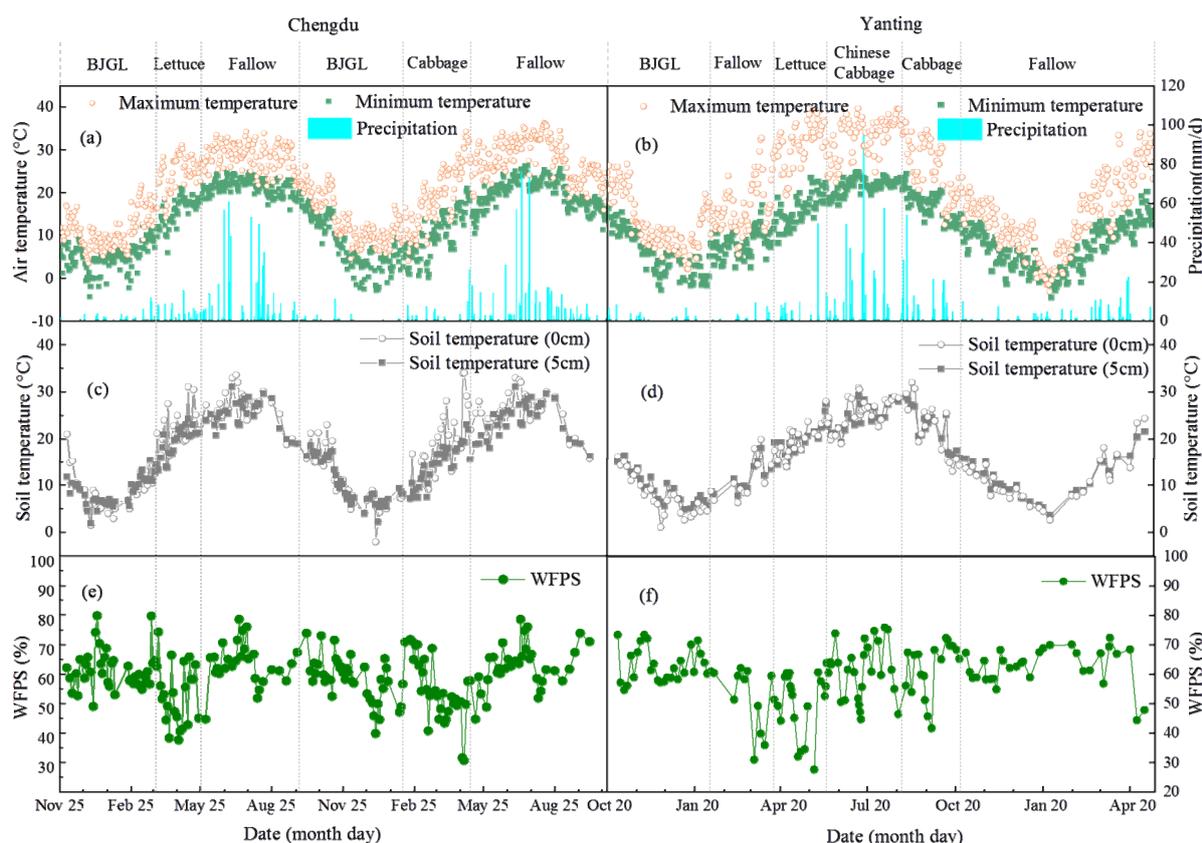


Fig. 1 Seasonal dynamics of maximum and minimum air temperature and precipitation (a-b), soil temperature in 0 and 5 cm depth (c-d), and mean water-filled pore space (WFPS) (e-f) at the Chengdu and Yanting sites. The data shown in panels (e-f) are the means of the fertilized and unfertilized treatments.

3.3 NO fluxes

At the CD site, soil NO emissions ranged from -4.7 to 83.1 $\mu\text{g N m}^{-2} \text{h}^{-1}$ (mean: 20.7 $\mu\text{g N m}^{-2} \text{h}^{-1}$) for the N fertilization treatment and from -13.3 to 63.8 $\mu\text{g N m}^{-2} \text{h}^{-1}$ (mean: 2.7 $\mu\text{g N m}^{-2} \text{h}^{-1}$) for the control (Fig. 2). The pluses of NO emission were observed within 2 weeks following N fertilization and tillage events, which accounted for approximately 88.3% of seasonal cumulative NO emissions for the N fertilization treatment. The seasonal cumulative NO emissions were 0.48 kg N ha⁻¹ for the fertilized treatment and 0.06 kg N ha⁻¹ for the control, with the average EF_d of 0.21% (Table 3).

At the YT site, soil NO emissions ranged from -15.17 to 173.62 $\mu\text{g N m}^{-2} \text{h}^{-1}$ (mean: 8.56 $\mu\text{g N m}^{-2} \text{h}^{-1}$) for the N fertilization treatment and from -7.10 to 32.57 $\mu\text{g N m}^{-2} \text{h}^{-1}$ (mean: 0.73 $\mu\text{g N m}^{-2} \text{h}^{-1}$) for the control (Fig. 3). The seasonal cumulative NO fluxes ranged from 0.01 to 0.41 kg N ha⁻¹ (mean: 0.13 kg N ha⁻¹) for the N fertilization treatment and from 0.01 to 0.02 kg N ha⁻¹ (mean: 0.02 kg N ha⁻¹) for the control.

The NO pluses in YT site mainly occurred in the vegetable growing season following N fertilization and soil tillage, and contributed approximately 89.8% of annual NO fluxes. The annual NO fluxes were 0.75 kg N ha⁻¹ and 0.08 kg N ha⁻¹ for the N fertilization treatment and control, respectively. The EF_d of NO ranged from 0.05% to 0.27% (mean: 0.12%).

3.4 CH₄ fluxes

At the CD site, soil CH₄ fluxes ranged from -345.4 to 247.2 $\mu\text{g C m}^{-2} \text{h}^{-1}$ (mean: -28.2 $\mu\text{g C m}^{-2} \text{h}^{-1}$) for the N fertilization treatment and from -318.8 to 184.1 $\mu\text{g C m}^{-2} \text{h}^{-1}$ (mean: -40.1 $\mu\text{g C m}^{-2} \text{h}^{-1}$) for the control (Fig. 2). The annual cumulative soil CH₄ fluxes ranged from -2.07 to -2.39 kg C ha⁻¹ yr⁻¹ (mean: -2.23 kg C ha⁻¹ yr⁻¹) for the N fertilization treatment and from -2.63 to -3.4 kg C ha⁻¹ yr⁻¹ (mean: -3.02 kg C ha⁻¹ yr⁻¹) for the control (Table 3), i.e. the N fertilization practice significantly decreased soil CH₄ uptake.

At the YT site, soil CH₄ fluxes ranged from -124.7 to 60.6 $\mu\text{g C m}^{-2} \text{h}^{-1}$ (mean: -14.3 $\mu\text{g C m}^{-2} \text{h}^{-1}$) for the

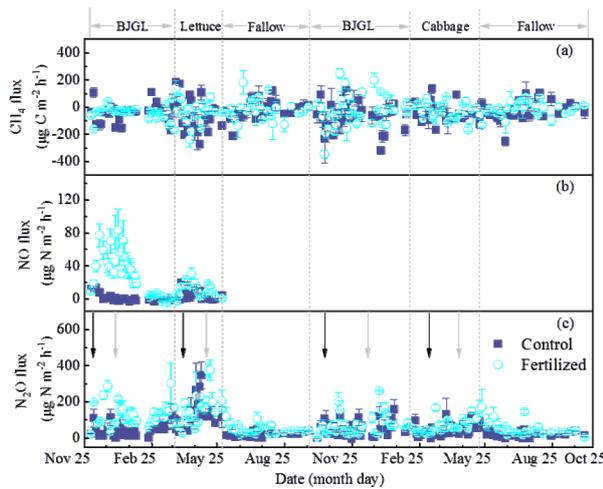


Fig. 2 Seasonal dynamics of (a) methane (CH₄), (b) nitric oxide (NO) and (c) nitrous oxide (N₂O) in the vegetable fields of Chengdu site from November 25, 2004 to October 25, 2006. Vertical bars indicate standard errors of three spatial replicates. The black and gray down arrows indicate the time of basal and topdressing fertilization, respectively. The NO emissions were not determined from May 26, 2005 to October 25, 2006 due to the instrument trouble, and the N₂O emissions from April 11 2005 to May 8 2006 were adopted and reprocessed from Yu et al. (2008).

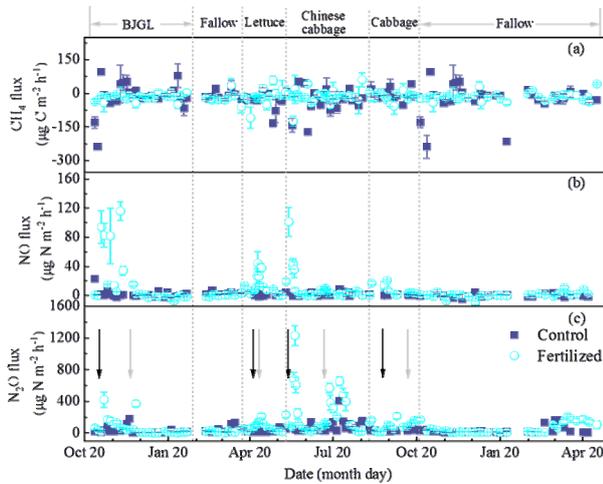


Fig. 3 Seasonal dynamics of (a) methane (CH₄), (b) nitric oxide (NO) and (c) nitrous oxide (N₂O) in the vegetable fields of Yanting from November 1, 2006 to May 6, 2008. Vertical bars indicate standard errors of three spatial replicates. The black and gray down arrows indicate the time of basal and topdressing fertilization, respectively. The N₂O emissions from October 25, 2006 to October 26, 2007 were adopted and reprocessed from Yu et al. (2012).

N fertilization treatment and ranged from -238.5 to 94.5 µg C m⁻² h⁻¹ (mean: -21.3 µg C m⁻² h⁻¹) for the control (Fig. 3). The annual cumulative soil CH₄ fluxes ranged from -1.02 to -1.63 kg C ha⁻¹yr⁻¹ (mean: -1.25

kg C ha⁻¹ yr⁻¹) for the N fertilization treatment and from -1.95 to -2.01 kg C ha⁻¹yr⁻¹ (mean: -1.97 kg C ha⁻¹ yr⁻¹) for the control (Table 3).

4 Discussion

4.1 N₂O and NO fluxes

Soil N₂O and NO emission peaks were mostly observed following N fertilization events (i.e. within 11 days for CD and 5 days for YT after N fertilization events) (Figs. 2 and 3), the alkaline soil in YT peaked faster is likely with increasing soil pH have more free NH₃ for ammonia oxidation and the generated nitrite and nitrate is advantageous to denitrification (Butterbach-Bahl et al. 2013). These temporal patterns of soil N₂O and NO emissions were in line with previous studies on conventional vegetable cultivation systems (e.g., Pang et al. 2009; Xie et al. 2019; Yao et al. 2015; Zhang et al. 2019).

It is noteworthy that N₂O emission pulses due to fertilization and precipitation events significantly contributed to annual cumulative N₂O fluxes from the subtropical conventional vegetable systems at both CD (69.5%) and YT (81.1%) sites, although the pulses occurred with a relatively short period (Figs. 2 and 3). Similarly, soil N₂O pulses contributed to 86.0% of seasonal N₂O fluxes in a conventional vegetable system in eastern China (Mei et al. 2011). Nevertheless, either fertilization or rainfall events induced significant soil N₂O pulses during the winter growing seasons (Figs. 2 and 3). This phenomenon is likely explained by the low soil temperature, which was positively correlated with soil N₂O emissions throughout the experiment (Fig. 4). Numerous studies have demonstrated that the N₂O emission related to soil nitrification and denitrification processes are highly temperature-dependent (Szukics et al. 2010). Thus, it highlights that further mitigation practices have to well consider the warm seasons (spring to autumn), particularly the “hot moments” of soil N₂O emissions due to fertilization and rainfall events for the subtropical conventional vegetable systems.

On average, the annual N₂O emissions for N fertilization treatment at the CD and YT sites were 6.34 and 7.71 kg N ha⁻¹, respectively, which fell within the range of 1.8-66.6 kg N ha⁻¹ for Chinese conventional vegetable systems (Deng et al. 2013; Mei

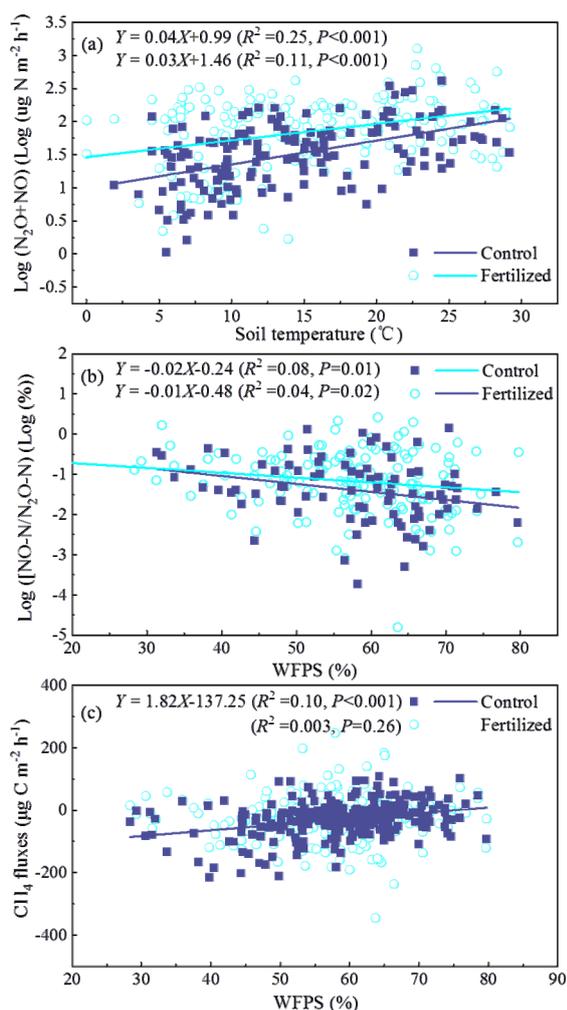


Fig. 4 Dependency of the sum of NO-N and N₂O-N fluxes on soil temperature (°C) (a). Dependency of the mass flux ratio of NO/N₂O (b) and CH₄ fluxes (c) on soil water-filled pore space (WFPS). Empty and solid dots denote the data from fertilized and control treatments in both sites.

et al. 2011; Xiong et al. 2006; Yao et al. 2015). Nevertheless, annual N₂O fluxes from the subtropical conventional vegetable systems in our study were lower than those in South China [Deng et al. (2012): 20.8 kg N ha⁻¹] and in North China Plain [Xie et al. (2019): 28.69 kg N ha⁻¹] while greater than those for cereal cropping systems in the same study region [e.g., Zhou et al. (2014): 1.73 kg N ha⁻¹ yr⁻¹ in the upland wheat-maize rotation system with annual N application rates of 280 kg N ha⁻¹ yr⁻¹; Zhou et al. (2018): 1.61 kg N ha⁻¹ yr⁻¹ in the paddy fields with annual N application rates of 250 kg N ha⁻¹ yr⁻¹]. The relatively low annual N₂O emissions in both sites might attributed to the following reasons: 1) the lower

fertilization rate compared with other vegetable systems (e.g., annual N rate > 1000 kg N ha⁻¹ at Wuxi, Jiangdu and Beijing, or seasonal N rate > 400 kg N ha⁻¹ at Zhangqiu and Nanjing, Table 3); 2) without carbon sources incorporation which might largely stimulate N₂O production through denitrification process (Butterbach-Bahl et al. 2013; Senbayram et al. 2018); 3) with a longer fallow period, and 4) higher soil pH (7.37-8.35) could decrease the N₂O/(N₂O+N₂) product ratio of denitrification and simultaneously increase the risk of soil NO₃⁻ losses which can be used as substrates for denitrification, thereby inhibiting N₂O emissions (Butterbach-Bahl et al. 2013; Pilegaard 2013; Van Cleemput & Samater 1995).

Similar patterns were also observed for soil NO emissions. Soil NO emission pulses due to fertilization and tillage events accounted for 88.3% and 89.8% of total NO fluxes at the CD and YT sites, respectively (Figs. 2 and 3), which also fell within the observed range (32% to 95%) in a four years field experiment on conventional vegetable systems (Mei et al. 2009). Nevertheless, annual soil NO fluxes (0.75 kg N ha⁻¹ yr⁻¹) in the present study were lower than the previous observations on the subtropical conventional vegetable systems in Tai-lake region (Mei et al. (2009): 1.1 to 11.9 kg N ha⁻¹ with an average of 5.7 kg N ha⁻¹, with annual N application rates of 1124 kg N ha⁻¹ yr⁻¹) as well as the global average of 9.8 kg N ha⁻¹ for vegetable systems (Liu et al. 2017). The relatively low soil NO fluxes in the present study might be due to the high soil pH that could decrease the production of NO+N₂O and their production ratio (Jiang et al. 2015; Wang et al. 2018b). Moreover, the high soil moisture conditions (e.g. the average of 59.6% WFPS at YT site) in the subtropical conventional vegetable systems might inhibit NO emission (Pilegaard 2013). In the present study, the average NO/N₂O ratios of 0.15 for the CD site and 0.13 for the YT site indicated that nitrification might be the main process for soil NO emissions from the subtropical conventional vegetable cultivation systems; because nitrification generally occurred predominantly when NO/N₂O > 0.11 (Meijide et al. 2007). Our findings were consistent with the previous studies that nitrification is usually the major NO production process in upland soils [e.g., as reviewed by Medinets et al. (2015)]. This is further evidenced by the negative correlations between the NO/N₂O ratio and soil WFPS (Fig. 4), that is because low soil WFPS conditions favor the nitrification process for NO production (Wu et al.

2017) while inhibiting NO reduction by denitrification (Bateman & Baggs 2005).

4.2 Direct N₂O and NO emission factors and background emissions

The lack of multi-site field measurements of direct N₂O and NO emission factors (EF_d) and background emissions for conventional vegetable systems likely led to the uncertainties in regional or global estimates of N₂O and NO emissions (Liu et al. 2017; Wang et al. 2011; Wang et al. 2018a). For example, case studies with simultaneous N₂O and NO determination in Asia vegetable cultivation systems were synthesized (Table 4), and results showed that the great variations in soil N₂O and NO fluxes were most likely due to differences in site-specific conditions, such as climate, soil type and management practice (Bouwman et al. 2002b; Butterbach-Bahl et al. 2013). Furthermore, as the gaseous product of soil N transformation, the exchanges of N₂O and NO between soil and atmosphere are closely interrelated (Yao et al. 2015). Therefore, it highlights the importance of simultaneous quantifications of N₂O and NO emissions on the basis of multi-site field measurements for most regional common practices. In the present study, the average N₂O EF_d values of 0.73% for the CD site and 0.68% for the YT site were in the lower range of lasted default EF_d value of 0.70-1.9% (mean: 1.3%) recommended by the IPCC (Hergoualc'h et al. 2019) for temperate/boreal wet climate. However, the present results were greater than that for the global average by a global meta-analysis [(Wang et al. 2011): 0.55%] while much lower than that for the subtropical conventional vegetable cultivation systems (from 1.20% to 5.00%) in southeastern China (Jia et al. 2012; Mei et al. 2011). The average NO EF_d value of 0.12% obtained in the present study was over ten times lower than that for the global average of 1.71% for conventional vegetable systems while comparable with that 0.15% for cereal cropping systems (0.15%) in the same study region (Xiao et al. 2018).

In the present study, background soil N₂O emissions for the subtropical conventional vegetable cultivation systems (3.42 to 3.62 kg N ha⁻¹ yr⁻¹) were two times higher than that of the average background soil N₂O for the Chinese vegetable cultivation systems

[(Wang et al. 2018a): 1.25 kg N ha⁻¹ yr⁻¹]. However, background soil NO emissions (mean: 0.08 kg N ha⁻¹ yr⁻¹) obtained in the present study were ten times lower than the global average of 0.97 kg N ha⁻¹ yr⁻¹ for vegetable cultivation systems (Liu et al. 2017). These phenomena suggest that background soil N₂O and NO emissions from different sites varied largely were probably ascribed to different soil properties (e.g., soil pH) and site-specific climate.

4.3 Soil CH₄ fluxes

In general, the subtropical conventional vegetable cultivation systems acted as weak sinks of atmospheric CH₄ on the annual scale at both experimental sites (Table 3), which were consistent with previous observations in the same study region (Zhou et al. 2014; Zhou et al. 2019). However, Jia et al. (2012) found that vegetable cultivation systems were net source of atmospheric CH₄, because the frequent irrigations could formulate soil anaerobic condition for CH₄ production. We also found that the CH₄ fluxes were positively correlated with WFPS ($p < 0.0001$) in the control treatment, which means the soil turn from CH₄ sink to CH₄ source with the increasing of soil moisture. However, no significant correlations were found between soil CH₄ fluxes and WFPS in the fertilized treatment in both sites.

Previous studies have found significant N fertilization effects on the magnitude of soil CH₄ uptakes, although its effects on soil CH₄ oxidation are complex and not well clarified so far (Le Mer & Roger 2001; Zhou et al. 2018). In the present study, N fertilizer applications significantly decreased soil CH₄ uptakes as compared with the control (Table 3), which were inconsistent with the previous study by (Hellebrand et al. 2003) that N fertilizer application enhanced the activity of methanotrophs thereby increasing soil CH₄ uptakes. Nevertheless, in line with our current study, the recent meta-analysis found that N fertilization practices significantly reduced soil CH₄ uptakes in particular if the N application rates exceeded the threshold of 100 kg N ha⁻¹ yr⁻¹ (Aronson & Helliker 2010). These phenomena could be explained by the competitive inhibition of methane monooxygenase by ammonium or the toxicity/osmotic effect of hydroxylamine and nitrite on methanotrophic bacteria due to N addition (Gulledge et al. 2004).

Table 4 Summary of soil pH, vegetable cultivars, fertilizer types (F_T), fertilizer N rates (F_{NR} kg N ha⁻¹), seasonal N₂O and NO fluxes (S_F kg N ha⁻¹), direct emission factors (EF_d) (%), background fluxes (S_B kg N ha⁻¹) and ratio of NO/N₂O from fertilized (R_F) and unfertilized (R_B) (background emission) vegetable fields in Asia, and only studies reporting with simultaneously N₂O and NO determination were included [include 6 open vegetable fields (1-6) and 2 greenhouse vegetable fields (7-8)].

Site	pH	Time	Vegetables	F _T	F _{NR}	S _F -N ₂ O	EF _d -N ₂ O	S _F -NO	EF _d -NO	S _B -N ₂ O	S _B -NO	R _F	R _B	
Tsukuba, Japan ¹	5.6	Sep 1999-Nov 1999	Cabbage	Urea	250	0.78	0.16	1.95	0.78	0.38	0.01	2.50	0.03	
Tsukuba, Japan ²	5.6	Sep 2000-Feb 2001	Chinese cabbage	Urea	250	0.56	0.18	1.00	0.40	0.12	-	1.78	-	
Wuxi, China ³	6.2	Oct 2003-Feb 2004	Green vegetables	M	68	1.88	1.03	10.00	14.60	2.30	0.10	5.32	0.04	
		Feb 2004-May 2004	Lettuce	Urea + M	499	0.93	0.36	11.20	2.21	4.98	0.16	12.06	0.03	
		May 2004-Jul 2004	Chinese cabbage	M	68	14.5	0.67	19.30	28.2	10.5	0.17	1.32	0.02	
		Jul 2003-Sep 2004	Chinese cabbage	Urea	232	0.54	0.40	2.08	2.08	0.80	1.66	0.24	3.85	0.14
		Sep 2004-Oct 2004	Green vegetables	Urea	193	0.15	0.20	1.99	0.98	0.48	0.09	0.09	13.27	0.19
		Oct 2004-Nov 2004	Green vegetables	Urea	232	0.25	0.32	2.57	1.09	0.91	0.04	0.04	10.28	0.04
		Sep 2004-Nov 2004	Radish	SC	130	1.97	-	0.10	0.01	0.01	-	0.09	0.05	-
		Nov 2004-May 2005	Vegetable rape	SM + CF	413	2.22	0.18	0.65	0.13	1.48	0.11	0.11	0.29	0.07
		May 2005-Sep 2005	Chili	RC + M	138	4.03	2.62	0.35	0.14	0.40	0.16	0.16	0.09	0.40
		Sep 2005-Nov 2005	Radish	Urea + SC	548	46.60	8.43	6.69	1.20	0.48	0.13	0.13	0.14	0.27
Jiangdu, China ⁴	7.9	Nov 2005-Mar 2006	Vegetable rape	RC + CF	638	0.56	0.06	0.42	0.05	0.26	0.08	0.75	0.31	
		Mar 2006-Aug 2006	Amaranth	M + SC	382	54.70	14.20	4.74	1.19	0.52	0.19	0.09	0.37	
		Aug 2006-Nov 2006	Radish	Urea + SC	397	10.20	2.49	5.02	1.24	0.29	0.10	0.49	0.34	
		Nov 2006-Jun 2007	Garlic	RC + CF	385	2.31	0.45	0.45	0.11	0.59	0.03	0.19	0.05	
		Jun 2007-Aug 2007	Amaranth	RC + CF	118	2.15	1.57	0.30	0.23	0.30	0.04	0.14	0.13	
		Aug 2007-Aug 2007	Fallow	Urea	153	2.71	1.71	1.08	0.67	0.10	0.10	0.05	0.40	0.50
		Aug 2007-Nov 2007	Radish	Urea + SC	418	11.00	2.56	1.39	0.32	0.27	0.04	0.04	0.13	0.15
		Nov 2007-Mar 2008	Garlic	RC+SC+CF	393	2.86	0.68	0.68	0.09	0.18	0.10	0.10	0.16	0.56
		May 2008-Aug 2008	Amaranth	RC + CF	201	1.61	0.73	1.04	0.48	0.15	0.06	0.06	0.65	0.40
		Sep 2006-Dec 2006	Cabbage	Urea + M	271	0.48	0.18	0.18	0.96	-	0.24	5.89	-	
Jiaxing, China ⁵	6.1	Sep 2006-Dec 2006	Garlic	Urea + M	267	0.73	0.27	30.20	11.20	-	-	41.40	-	
		Sep 2006-Dec 2006	Radish	Urea + M	264	0.36	0.14	6.40	2.34	-	-	17.80	-	
Zhangqiu, China ⁶	8.1	Jun 2009-Nov 2009	welsh onion	CF + M	466	2.92	0.53	0.76	0.11	0.44	0.24	0.26	0.55	
		Jun 2010-Nov 2010	welsh onion	CF + M	487	2.76	0.44	1.38	0.22	0.63	0.33	0.50	0.52	
Nanjing, China ⁷	5.4	Aug 2013-Mar 2014	Tomato	CF	400	5.20	1.03	4.96	1.08	1.09	0.63	0.95	0.58	
		Mar 2014-May 2014	Chinese cabbage	CF	150	2.09	0.98	1.55	0.89	0.62	0.21	0.74	0.34	
Beijing, China ⁸	7.6	May 2014-Aug 2014	Green soybean	CF	90	6.97	4.54	2.62	2.45	2.71	0.42	0.38	0.15	
		Mar 2016-Jun 2016	Cucumber	CF+M	1200	28.70	1.78	0.86	0.08	7.32	0.32	0.03	0.04	

Note: ¹(Hou & Tsuruta 2003); ²(Cheng et al. 2002); ³(Deng et al. 2012); ⁴(Mei et al. 2009, Mei et al. 2011); ⁵(Pang et al. 2009); ⁶(Yao et al. 2017); ⁷(Zhang et al. 2019); ⁸(Xie et al. 2019). M: manure; RC: rapeseed cake; SC: soybean cake; CF: compound fertilizer. - means values unreported in the studies.

5 Conclusion

The direct N₂O and NO emission factors in the subtropical conventional vegetable cultivation systems were in the lower range of the latest default EF_a value of 0.70-1.9% recommended by (Hergoualc'h et al. 2019). The annual background N₂O emissions for subtropical conventional vegetable cultivation systems are higher than those for the global average, while the background NO emissions are negligible. Soil pH, temperature and moisture conditions are the key regulators of soil N₂O and NO emission and contributed to the corresponding temporal variations of soil N₂O and NO emissions. Soil acted as sink of atmospheric CH₄ while N fertilizer application inhibited soil CH₄ uptakes. Nevertheless, further field

studies, particularly for alkaline soils, are needed to improve the accuracy of the regional and global N₂O, NO and CH₄ inventories and to propose mitigation practices for the subtropical conventional vegetable cultivation systems.

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