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Baoling Mei
Chinese Academy of Sciences

Xunhua Zheng
Chinese Academy of Sciences

Baohua Xie
Chinese Academy of Sciences

Haibo Dong
Chinese Academy of Sciences

Zhisheng Yao
Chinese Academy of Sciences

See next page for additional authors

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Authors

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Characteristics of multiple-year nitrous oxide emissions from conventional vegetable fields in southeastern China

Baoling Mei,[1,2] Xunhua Zheng,[1] Baohua Xie,[1] Haibo Dong,[1] Zhisheng Yao,[1] Chunyan Liu,[1] Zaixing Zhou,[1] Rui Wang,[1] Jia Deng,[1] and Jianguo Zhu[3]

[1]   The annual and interannual characteristics of nitrous oxide (N_2O) emissions from conventional vegetable fields are poorly understood. We carried out 4 year measurements of N_2O fluxes from a conventional vegetable cultivation area in the Yangtze River delta. Under fertilized conditions subject to farming practices, approximately 86% of the annual total N_2O release occurred following fertilization events. The direct emission factors (EF_d) of the 12 individual vegetable seasons investigated ranged from 0.06 to 14.20%, with a mean of 3.09% and a coefficient of variation (CV) of 142%. The annual EF_d varied from 0.59 to 4.98%, with a mean of 2.88% and an interannual CV of 74%. The mean value is much larger than the latest default value (1.00%) of the Intergovernmental Panel on Climate Change. Occasional application of lagoon-stored manure slurry coupled with other nitrogen fertilizers, or basal nitrogen addition immediately followed by heavy rainfall, accounted for a substantial portion of the large EF_ds observed in warm seasons. The large CVs suggest that the emission factors obtained from short-term observations that poorly represent seasonality and/or interannual variability will inevitably yield large uncertainties in inventory estimation. The results of this study indicate that conventional vegetable fields associated with intensive nitrogen addition, as well as occasional applications of manure slurry, may substantially account for regional N_2O emissions. However, this conclusion needs to be further confirmed through studies at multiple field sites. Moreover, further experimental studies are needed to test the mitigation options suggested by this study for N_2O emissions from open vegetable fields.

1.   Introduction

[2]   Nitrous oxide (N_2O) is a particularly important member of the nitrogen oxides because it is both a greenhouse gas with a long residence time in the atmosphere and an important source of stratospheric nitric oxide (NO), and the latter role is essential in ozone layer chemistry. The atmospheric concentration of N_2O has been increasing at a rate of 0.2–0.3% per year [*Intergovernmental Panel on Climate Change (IPCC)*, 2007]. Cultivated soils, which are major biological sources of N_2O, emit the gas mainly through microbial nitri-fication and denitrification. Globally, approximately 2.8 Tg N y^{-1} is currently released from cultivated soils as N_2O [*IPCC*, 2007], and nitrogen (N) fertilizers are the dominant sources of N_2O emissions from cultivated soils [*Mosier and Kroeze*, 2000; *Stehfest and Bouwman*, 2006].

[3]   To meet the food requirements of a rapidly increasing world population, especially in developing nations, the use of N fertilizers is essential. Global consumption of N fertilizers increased at a rate of 6–7% yr^{-1} in the 1990s [*Food and Agriculture Organization (FAO)*, 1998], and the forecast for world N fertilizer demand shows an increase at an annual rate of approximately 1.4% (which is an overall increase of 7.3 million tons per year) through 2011/2012; approximately 69% of this growth will take place in Asia [*FAO*, 2008]. China's large, growing population is associated with an immense agricultural demand. As a result, the country currently consumes almost 1/3 of the world's N fertilizers (http://faostat.fao.org/default.aspx). Of the N fertilizer consumed in China, a considerable proportion (e.g., approximately 17% in the 1990s) has been applied to vegetable farms [*Zheng et al.*, 2004]. Vegetable production in China

[1]State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China.
[2]College of Environment and Resources, Inner Mongolia University, Huhhot, China.
[3]State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing, China.

has increased rapidly over the past two decades. By the early 2000s, for instance, the vegetable cultivation area had increased to approximately 19% of the total crop cultivation area of China from representing less than 10% in the 1980s [*FAO*, 2004]. Furthermore, vegetable farms are usually treated with greater amounts of N fertilizers than fields cultivated with grain crops. Nitrogen application rates for individual vegetable growing seasons are usually 300 to 700 kg N ha^{-1} [e.g., *Li and Wang*, 2007] and even more in some cases [e.g., *Ju et al.*, 2004; *He et al.*, 2009], but the rate is only 150 to 300 kg N ha^{-1} for nonvegetable crops [e.g., *Ju et al.*, 2009]. The increased use of N fertilizers has stimulated and will most likely continue to stimulate intensive N$_2$O emissions from vegetable fields. Therefore, the development of mitigation strategies for controlling N$_2$O emissions while sustaining vegetable yields is urgent. Understanding the characteristics and quantifying the magnitude of N$_2$O emissions from vegetable fields will provide a scientific basis for developing mitigation options.

[4] There have been a number of field studies on N$_2$O emissions from vegetable fields all over the world [*Smith et al.*, 1998; *van der Weerden et al.*, 2000; *Ruser et al.*, 2001; *Cheng et al.*, 2002; *Gattinger et al.*, 2002; *Flessa et al.*, 2002; *Kusa et al.*, 2002; *Hou and Tsuruta*, 2003; *Yang et al.*, 2005; *Burger et al.*, 2005; *Xiong et al.*, 2006; *Cao et al.*, 2006a, 2006b; *Hosono et al.*, 2006; *Vallejo et al.*, 2006; *Toma et al.*, 2007; *He et al.*, 2007, 2009; *Vermeulen and Mosquera*, 2009; *Pang et al.*, 2009; *Lin et al.*, 2010]. However, only a few of these studies were based on multiyear field measurements, and approximately half of the previous studies with year-round measurements were carried out in vegetable fields within greenhouses [*Xiong et al.*, 2006; *Hosono et al.*, 2006; *He et al.*, 2007, 2009]. The study with the longest field measurements (6 years) on this topic was conducted at a study site with gray lowland soil in Hokkaido, Japan. In this previous study, only onion was cultivated, and observations of N$_2$O emissions were conducted only during the growing seasons [*Kusa et al.*, 2002]. However, measurements made exclusively in growing seasons may underestimate total annual emissions, as strong N$_2$O emissions may sometimes occur during fallow periods. We found eight reports in the literature on N$_2$O emissions related to vegetable cultivation in China. Of these Chinese studies, three were based on measurements conducted in the fields within greenhouses [*Xiong et al.*, 2006; *He et al.*, 2007, 2009]; one employed outdoor potting experiments [*Yang et al.*, 2005]; and four were conducted using observations made in open vegetable fields [*Cao et al.*, 2006a, 2006b; *Pang et al.*, 2009; *Lin et al.*, 2010]. Out of the four reports on N$_2$O emissions from open vegetable fields, the field measurements of three studies were conducted over short periods of three to seven months [*Cao et al.*, 2006a, 2006b; *Pang et al.*, 2009], and only one study was conducted over a long period of four years [*Lin et al.*, 2010]. The N$_2$O fluxes measured within periods much shorter than one year may poorly represent the annual emissions. In the single multiyear measurement study, observations of gas fluxes were monthly conducted [*Lin et al.*, 2010]. Such a low frequency of observations may not represent high temporal variability. Especially explosive N$_2$O pulses may be missed. As a result, enormous uncertainty may be yielded for emission factors, which are key parameters for inventory

compilation [*IPCC*, 2006]. Additionally, vegetable growing conditions are quite different between greenhouses and open fields. Thus, emissions measured inside greenhouses may differ considerably from those of open fields. Moreover, N$_2$O emissions from vegetable fields are regulated not only by the dose of N fertilizers applied but also by environmental conditions, such as soil temperature. For instance, 2 year observations conducted in a tomato field inside a greenhouse in Shandong province showed that 0.27–0.30% of fertilizer N was lost through N$_2$O emissions [*He et al.*, 2009], but *Cao et al.* [2006b] observed much higher loss rates of 2.62–4.92% in open vegetable fields in Nanjing, China. Many factors, such as the time of observations, soil properties, and management practices, may have accounted for the tremendous difference in the results of these two studies, and these are factors that were poorly addressed by the authors.

[5] To better understand the large temporal and/or spatial variability observed in N$_2$O emissions from vegetable fields and its essential driving forces, intensive year-round and multiyear measurements under open conditions with conventional management practices are required. To meet these requirements, we launched a study using 4 year simultaneous measurements of N$_2$O and NO emissions from open vegetable fields in the Yangtze River delta under the local conventional management regime. Our NO emission results were reported in a separate recent publication [*Mei et al.*, 2009]. In this report, we will pay more attention to the results of N$_2$O emissions. Furthermore, the NO emissions reported by *Mei et al.* [2009] and N$_2$O emissions reported here will be synthetically compared and discussed.

2. Materials and Methods

2.1. Experimental Site and Field Treatments

[6] We conducted experiments in the Yangtze River delta from 1 September 2004 through 5 October 2008. Before we began our experiments, the soil had been conventionally cultivated with upland vegetables for approximately 20 years. Prior to vegetable cultivation, the soil was a paddy (specifically, a Shajiang Hapli-Stagnic Anthrosol) cultivated with annual rice-wheat or rice-rapeseed rotation systems. In the present study, two fertilization treatments (with and without addition of N fertilizer) were applied, and three replicate field plots were randomly set for each treatment. The details of the experimental sites, soil properties, field management practices, meteorological conditions (daily precipitation and daily mean air temperature) and soil conditions (daily soil moisture in water-filled pore space and daily soil temperature) for the entire experimental period were described in our recent publication on NO emissions [*Mei et al.*, 2009]. Our experiments spanned four full-year periods and included thirteen vegetable growing seasons and a short fallow period, which were denoted as P1 through P14 in Table 1. The exact time spans for these periods are provided in the footnotes of this table. For the fertilization treatments both with and without N addition, we performed our experiments in a field managed by a local farmer following the conventional cultivation practices in the region. All management practices of both field treatments during our investigation period were carried out by the local farmer, such as choosing the vegetable species, fertilization schedules and doses, and watering schedules. The experimental fields were cultivated with radishes

Table 1. Crop Species, Fertilization Events, and Fertilization Rates[a]

| | Vegetable | Fertilization | | | |
		Date	Type	Fertilizers	Rate[d]
P1	Radish (*R. sativus*)[b]	3 Sep 2004	Basal	Soybean cake	130
P2	Vegetable rape (*B. napus*)[c]	29 Nov 2004	Basal	Swine manure, CF	205
		28 Feb 2005	TD	Urea	208
P3	Chili (*C. frutescens*)[c]	29 May 2005	Basal	Rapeseed residues, silkworm manure	138
P4	Radish[b]	2 Sep 2005	Basal	Urea, soybean cake	548
P5	Vegetable rape[c]	27 Nov 2005	Basal	Rapeseed cake, CF	326
		16 Jan 2006	TD	Urea	312
P6	Amaranth (*A. mangostanus*)[b]	27 Mar 2006	Basal	Human manure slurry, soybean cake	382
P7	Radish[b]	17 Aug 2006	Basal	Urea, soybean cake	397
P8	Garlic (*A. satvum*)[c]	4 Nov 2006	Basal	Rapeseed cake, CF	177
		4 Mar 2007	TD	Urea	208
P9	Amaranth[b]	2 Jun 2007	Basal	Rapeseed cake, CF	118
P10	Fallow	7 Aug 2007	Basal	Urea	153
P11	Radish[b]	21 Aug 2007	Basal	Urea, soybean cake, CF	418
P12	Garlic[c]	12 Nov 2007	Basal	Rapeseed and soybean cake, CF	144
		11 Mar 2008	TD	Urea	249
P13	Amaranth[b]	29 May 2008	Basal	Rapeseed cake, CF	201
P14	Radish[b]	20 Aug 2008	Basal	Rapeseed cake, CF	281

[a]The same plant species were grown in both the fertilized and unfertilized plots. Radish (*Raphanus sativus*) was cultivated during P1 (3 Sep ~ 28 Nov 2004), P4 (2 Sep ~ 26 Nov 2005), P7 (10 Aug ~ 3 Nov 2006), P11 (21 Aug ~ 11 Nov 2007) and P14 (20 Aug ~ 5 Oct 2008, not harvested); vegetable rape (*Brassica napus*) during P2 (29 Nov 2004 ~ 22 May 2005) and P5 (27 Nov 2005 ~ 26 Mar 2006); amaranth (*Amaranthus mangostanus*) in P6 (27 Mar ~ 9 Aug 2006), P9 (2 Jun ~ 3 Aug 2007) and P13 (25 May ~ 19 Aug 2008); chili (*Capsicum frutescens*) during P3 (23 May ~ 1 Sep 2005); and garlic (*Allium astvum*) during P8 (4 Nov 2006 ~ 1 Jun 2007) and P12 (12 Nov 2007 ~ 24 May 2008). The soil was fallow during P10 (3 Aug ~ 20 Aug 2007). Basal, amended prior to vegetable sowing/transplanting. TD, top dressing. CF, compound fertilizer, which is a mixture of (NH$_4$)H$_2$PO$_4$ and KCl, with N:P$_2$O$_5$:K$_2$O = 15%:15%:15%.
[b]Seeds were directly sown in the fields.
[c]Seedlings were grown outside and were transplanted into the field.
[d]In kg N ha^{-1}.

(*Raphanus sativus*) in P1, P4, P7, P11, and P14, vegetable rape (*Brassica napus*) in P2 and P5, amaranth (*Amaranthus mangostanus*) in P6, P9, and P13, chili (*Capsicum frutescens*) in P3, and, garlic (*Allium satvum*) in P8 and P12. The soil was fallow in P10. Seeds of radishes and amaranth were directly sown in the experimental plots, while seedlings of the remaining vegetable crops were grown elsewhere and were then transplanted into the experimental fields. The soil was manually watered if there was no rainfall within a few days after vegetable sowing or transplanting. Each plot was annually irrigated with 6, 20, 19, and 25 mm of water during the four investigation years, respectively. Soon after the harvest of a vegetable crop, the soil was tilled to a depth of approximately 12–15 cm. At the same time, basal fertilizers were incorporated into the soil, and the next type of vegetable crop was then immediately sown or transplanted at the site. Organic manure and chemical fertilizers were applied to the plots in the fertilized treatments at rates of 1,074–1,312 (with a mean of 1,195) kg N ha^{-1} yr^{-1} (Tables 2 and 3). In the fertilized plots, organic manure, compound fertilizers and/or urea were basally applied independently or in combination for all crops, while urea was additionally top-dressed for vegetable rape and garlic. No fertilizer containing N was added in P2 through P14 to the plots treated without N addition, and only organic manure was basally applied at an amount of approximately 130 kg N ha^{-1} in P1 to all plots of both field treatments. In P6, human manure slurry from an indoor lagoon of a local farmhouse, in combination with soybean cake, was basally applied. The detailed schedules, fertilizer types and doses for individual fertilization events of P1 to P14 are described in Table 1. The N fertilization rates for all individual periods are summarized in Table 2, and the annual N addition rates are listed in Table 3.

2.2. Nitrous Oxide Flux Measurements

[7] Measurements of N$_2$O fluxes were initiated on 21 September 2004 (18 days after the basal application of organic manure and radish sowing that followed the tillage occurring 2 days previously) and continued until the end of the investigation period, spanning 1,475 days. At the center of each replicate plot, one miniplot was permanently defined for simultaneous measurements of N$_2$O and NO fluxes. The N$_2$O and NO fluxes were measured by sampling parallel air samples with an opaque static chamber, and the samples for N$_2$O were analyzed with a gas chromatograph (GC) equipped with an electronic capture detector (ECD) [*Wang and Wang*, 2003; *Zheng et al.*, 2008]. A stainless steel base collar (0.5 m × 0.5 m) with a groove on the top, which could be filled with water, was inserted 10 cm into the soil and remained there during the entire investigation period. For simultaneous measurement of N$_2$O and NO fluxes, a portable stainless steel chamber (0.5 or 1.0 m high, depending on the height of the vegetable plants) that exactly fit the groove was mounted onto the base collar. A gas-tight seal was ensured by filling the groove with water. Gas samples from the headspace for N$_2$O measurement were taken with a 60 mL gas-tight plastic syringe via a Teflon tube that was connected to a three-port valve. A second tube (Φ1 cm × 10 m) was installed on another side of the stainless steel chamber to balance the air pressure between the inside and outside conditions of the chamber. Two ventilators driven by 12 VDC were installed inside the top of the chamber to avoid the formation of gas concentration gradients. At each miniplot, simultaneous N$_2$O and NO fluxes were usually measured at intervals of 2–3 days by sampling between 09:00 and 11:00 local standard time (LST). However, immediately following events of rainfall, watering, fertilization or tillage,

Table 2. Total Amount of N₂O Emissions (N_2O_{ST}), Average Soil Moisture (M_{sa}), Average Soil Temperature (at 5 cm; T_{sa}), Amount of Applied Nitrogen Fertilizers (F_N), and Direct Emission Factors (EF_d) for Individual Vegetable Seasons/Periods in 2004–2008[a]

Period	F_N (t N ha⁻¹)	T_{sa} (°C) Mean	T_{sa} (°C) SD	M_{sa} (%WFPS) Mean	M_{sa} (%WFPS) SD	N_2O_{ST} (kg N ha⁻¹) N Applied Mean	N Applied SE	No N Applied Mean	No N Applied SE	EF_d[b] (%) Mean	EF_d[b] (%) SE
P1	0.130	18.7	3.2	59	8	1.97	0.43				
P2	0.412	8.6	5.9	61	12	2.22	0.73	1.48	0.52	0.18	0.22
P3	0.138	26.4	3.4	62	16	4.03	1.33	0.40	0.08	2.62	0.96
P4	0.548	20.6	5.4	65	7	46.64	12.28	0.48	0.25	8.43	2.24
P5	0.534	6.9	4.8	56	18	0.56	0.04	0.26	0.13	0.06	0.03
P6	0.382	21.3	5.2	56	14	54.70	2.89	0.52	0.21	14.20	0.76
P7	0.396	23.3	5.5	55	11	10.15	2.66	0.29	0.09	2.49	0.67
P8	0.385	8.4	5.8	56	14	2.31	0.05	0.59	0.09	0.45	0.03
P9	0.118	24.8	2.5	65	18	2.15	0.53	0.30	0.05	1.57	0.45
P10	0.153	28.3	2	62	11	2.71	0.55	0.10	0.01	1.71	0.36
P11	0.418	20.1	5.4	66	09	10.99	1.13	0.27	0.04	2.56	0.27
P12	0.394	9.5	6.9	54	10	2.86	0.60	0.18	0.04	0.68	0.15
P13	0.201	25.4	2.9	57	13	1.61	0.29	0.15	0.04	0.73	0.14
P14	0.281	24.2	1.6	65	13	2.96	0.42	0.12	0.03	1.01	0.15
Mean[c]						12.57	5.90	0.57	0.17	3.09	1.34
SD[c]						19.32		0.39		4.39	
CV (%)[c]						154		67		142	

[a]Definitions of P1 through P14 are found in Table 1. SD is standard deviation, and SE is standard error of three (but six for P1 with N applied) replicate plots.

[b]The direct emission factor was not determined in P1, as the measurements began three weeks after basal fertilization, and thus, intensive emissions due to N application might have been missed.

[c]P1, P10, and P14 are not included in the statistics.

daily measurements were conducted for 1–2 weeks. To determine N₂O fluxes, four (before 15 June 2005) or five air samples were taken from the chamber headspace during a period of 18 (in a growing or warm season) or 32 min (in a cold winter season or fallow period) at intervals of approximately 6 or 8 min. The air temperature inside the chamber and the air pressure of the ambient atmosphere were simultaneously recorded.

[8] The gas samples for N₂O measurement were stored in syringes for at most 10 h before they were analyzed with a GC-ECD instrument. The samples collected before 6 September 2005 were analyzed using the GC-ECD DN method defined by *Zheng et al.* [2008]. The samples collected from 6 September 2005 through 17 December 2006 were analyzed with the GC-ECD DN-Ascarite method described by *Zheng et al.* [2008], and those samples collected later were analyzed with the GC-ECD DN-CO₂ method defined by *Zheng et al.* [2008] and *Wang et al.*

[2010]. The DN-Ascarite and DN-CO₂ methods used dinitrogen (N₂) as the carrier gas, similar to the DN method. However, an ascarite (a type of sodium-hydroxide-coated silica) filter was additionally adopted in the DN-Ascarite method to remove carbon dioxide (CO₂) from the air samples [e.g., *Butterbach-Bahl et al.*, 1997], and a high concentration of CO₂ in N₂ (N₂:CO₂ = 90:10) was directly introduced into the ECD cell to buffer the unexpected effects of CO₂ in the samples on the N₂O signals [*Zheng et al.*, 2008; *Wang et al.*, 2010]. The DN-CO₂ method was adopted as a substitute for the DN-Ascarite method because it was more stable for N₂O analysis and more convenient for routine maintenance compared with DN-Ascarite [*Zheng et al.*, 2008; *Wang et al.*, 2010]. Further details on the chamber operation and sampling procedures are provided by *Zheng et al.* [2008] and *Mei et al.* [2009].

[9] The N₂O mixing ratios of the samples were calibrated with reference gases (0.3 to 1.0 μmol mol⁻¹ N₂O in dry air)

Table 3. Annual Doses of N Fertilizer Added (F_N), Total Water Input (Rainfall + Watering), Mean Air Temperature, Total Nitrous Oxide (N₂O) Release, and Direct N₂O Emission Factor (EF_d)

Period[a]	F_N (in t N ha⁻¹ yr⁻¹)	Rainfall + Watering (mm)	Air Temperature (°C)	N₂O releases (kg N ha⁻¹ yr⁻¹) Fertilized Mean	Fertilized SE	Unfertilized[b] Mean	Unfertilized[b] SE	EF_d (%) Mean	EF_d (%) SE
Annual 1	1.098	905	15.2	51.6	10.2	4.14	1.29	4.32	0.93
Annual 2	1.312	1026	15.5	66.6	1.6	1.23	0.58	4.98	0.13
Annual 3	1.074	753	16.3	18.9	0.6	1.31	0.17	1.64	0.05
Annual 4	1.294	873	15.4	8.2	0.6	0.51	0.03	0.59	0.04
Mean	1.195	889	15.6	36.3	16.0	1.81	1.00	2.88	1.24
SD	0.126	112	0.5	27.7		1.73		2.14	
CV (%)	11	13	3	76		96		74	

[a]Each period spanned from 21 September through 20 September of the following year.

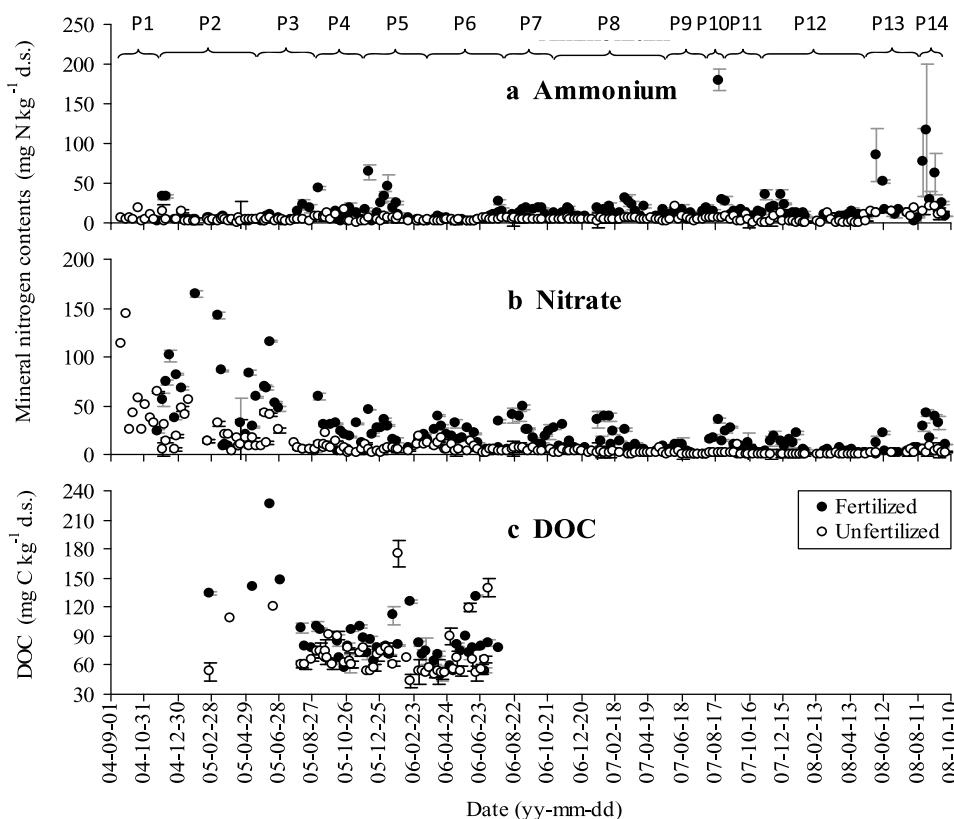[b]No fertilizer nitrogen was applied to the unfertilized vegetable fields.

Figure 1. Ammonium, nitrate and dissolvable organic carbon (DOC) levels in the soil of the cultivated layer (at 0–10 cm depth). Closed and open cycles denote the fertilized and unfertilized treatments (in terms of nitrogen addition), respectively. Definitions of P1 through P14 are found in Table 1.

from the National Standard Matter Center (Beijing, China). Each flux was calculated using the initial change rate of the N$_2$O concentrations within the chamber headspace, headspace volume, the soil surface area covered by the chamber, and N$_2$O density. The data on air pressure and chamber headspace air temperature were used to correct the N$_2$O density at 273 K and 1,013 hPa to the actual headspace air conditions in our flux calculations. The initial change rate was determined by nonlinear fitting of the four or five measured concentrations against the enclosure time [*Kroon et al.*, 2008]. Only the relationship of the measured N$_2$O concentrations with enclosure time was statistically significant at $p < 0.05$, and the initial change rate was accepted to yield a valid flux. The flux measurement was regarded as null whenever the relationship was not statistically significant. We compared the values of R^2 and the fluxes between the linear and nonlinear regressions to check the nonlinearity in the gas concentrations measured from the chambers. Only when both values of the significant nonlinear regression ($p < 0.05$) were higher than those of the significant linear regression ($p < 0.05$) was the flux calculated with the initial change rate determined with the significant nonlinear curve accepted. Otherwise, the flux calculated with the significant linear regression was accepted. For the DN-Ascarite and DN-CO$_2$ methods, the precisions for the analysis of N$_2$O concentrations in ambient air were ensured to be <1% [*Zheng et al.*, 2008; *Wang et al.*, 2010]. Taking

into account the enclosure time of 32 min, these precisions resulted in flux detection limits of 7 and 14 μg N m^{-2} h^{-1} for the chamber heights of 0.5 and 1.0 m, respectively.

[10] Details on the simultaneous measurement of NO fluxes were described by *Mei et al.* [2009].

2.3. Auxiliary Measurements

[11] In addition to recording field management practices, such as the dates and input rates of fertilizers and the amount of water input, we also measured soil temperature (at 5 cm), soil moisture (at 0–6 cm), ambient air temperature, precipitation and levels of ammonium (NH$_4^+$), nitrate (NO$_3^-$) and water dissolvable organic carbon (DOC) in the soils of the cultivated layer (0–10 cm). Daily precipitation and hourly air temperature were recorded by an automatic climate station located nearby (approximately 200 m away). The soil temperature in the direct vicinity of the chamber bases was measured daily using a thermocouple. Simultaneously, the ratio of the soil water content by volume was measured using a portable frequency domain reflector sensor (RDS Technology Co., Ltd Jiangsu, Nanjing, China). Soil moisture in the water-filled pore space (WFPS) was determined using the measured volumetric water content and soil porosity as 51% [*Mei et al.*, 2009]. Soil (0–10 cm) samples were collected weekly using a 3 cm diameter gauge auger for NH$_4^+$, NO$_3^-$ and DOC analysis. To measure NH$_4^+$ and NO$_3^-$ levels, fresh soil samples were extracted with a 0.01 mol L^{-1} CaCl$_2$
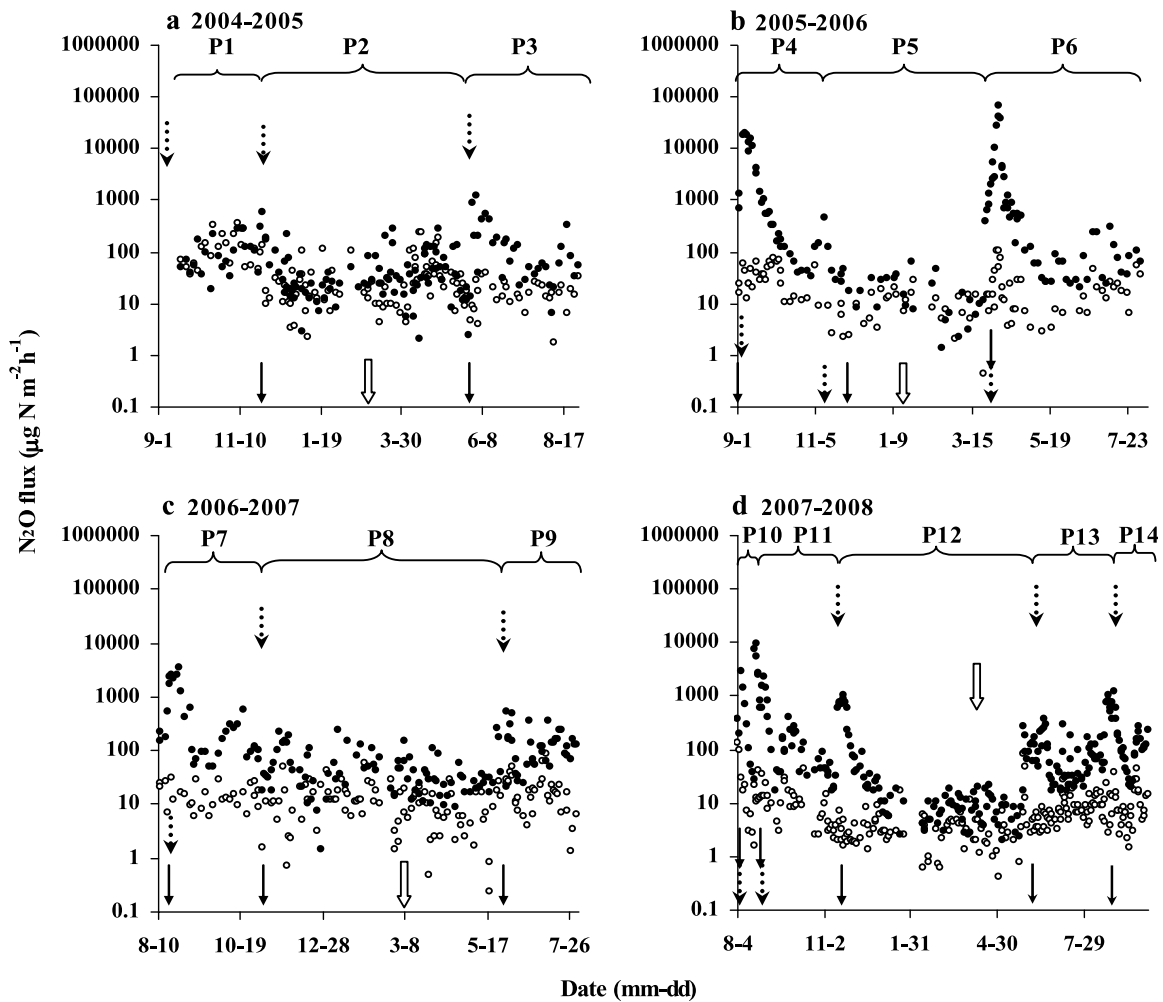
Figure 2. Nitrous oxide (N₂O) fluxes during the entire investigated period. Closed and open cycles denote the fluxes from the fertilized and unfertilized plots, respectively. Solid and open arrows indicate the dates of basal fertilization and urea top dressings, respectively, while dashed arrows indicate the tillage dates. Each datum is the mean of three replicates, with the error not shown.

solution (soil:water = 1:10) by shaking them for 1 h. The extracts were frozen at −18°C and thawed overnight at 4°C before being analyzed with an automatic nitrogen analyzer (AA2, BRAN & Lubbe, Nordstedt, Germany). Measurement of DOC was conducted from late February in 2005 to late July in 2006. Fresh soil samples were frozen at −18°C and thawed overnight at 4°C before extraction with deionized water (soil:water = 1:5) [e.g., *Lu*, 2000]. The extracts were immediately analyzed for DOC with a Multi N/C 3000 analyzer (Analytic Jena AG, Germany).

2.4. Data Analysis and Statistics

[12] As comparison studies [*Zheng et al.*, 2008; *Wang et al.*, 2010] have concluded, there are no obvious difference in the N₂O fluxes determined by the GC-ECD methods of DN-Ascarite, DN-CO₂, or AM, which uses an argon-methane mixture as the carrier gas, whereas the DN method results in significantly higher fluxes (the bias is especially problematic when opaque static chambers are used for gas sample collection). Based on a long-term comparison involving four field treatments (two of which were used in

this study) that lasted for approximately 1.5 years, *Zheng et al.* [2008] proposed an approach to correct the biases for the N₂O fluxes yielded by the DN method. Following that approach, we added 72, 22, −5, −38, and −64 μg N m^{-2} h^{-1} to the DN fluxes of <−30, −30 ~ 0, 0 ~ 30, 30 ~ 100, and 100 ~ 200 μg N m^{-2} h^{-1}, respectively, to correct the biases.

[13] The single N₂O flux measured on a day was directly extrapolated to the daily total emissions. Emissions on the days without measurements were estimated using the arithmetic mean of the two temporally adjacent observations. By summing the daily fluxes, we obtained seasonal/periodical or annual cumulative emissions. This approach to estimate seasonal/periodical or annual total emissions was based on two assumptions: (1) the single flux measured during 09:00–11:00 LST was representative of the daily mean, and (2) the missing daily N₂O emissions could be represented by the arithmetic mean flux of the two temporally adjacent observations [*Yao et al.*, 2009].

[14] The total nitrous oxide released from a given crop cultivation area is composed of both background and direct
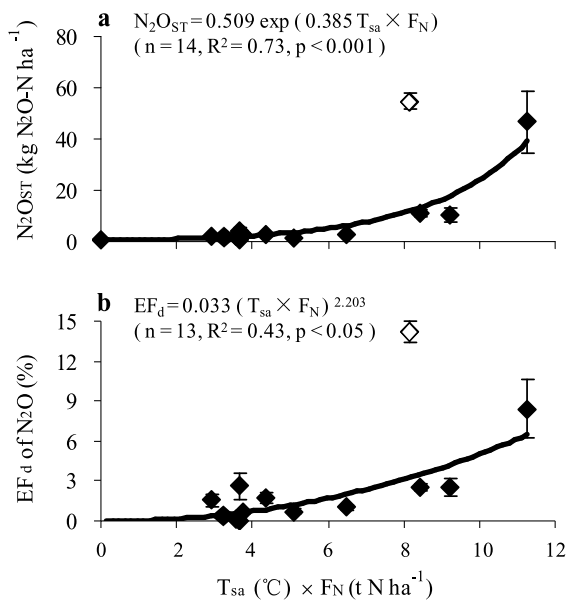
Figure 3. Interactive effects of the seasonal/periodical average soil (5 cm) temperature (T_{sa}) and fertilizer nitrogen addition rate (F_N) on (a) the seasonal/periodical cumulative nitrous oxide (N_2O) emissions (N_2O_{ST}) and (b) direct N_2O emission factors (EF_d). The given data are means of replicates (three observations for N_2O_{ST} of the fertilized treatment and 13 observations for the unfertilized plots). Vertical bars indicate standard errors. The open diamond symbol is for P4, in which human manure slurry was exclusively applied.

emissions. According to concept of the IPCC, direct emissions are derived from nitrogenous fertilizer(s) applied in the current season/period or year [*IPCC*, 1997, 2006]. Background emissions are derived from nitrogen sources other than the fertilizer(s) applied in the current season/period or year [*Bouwman*, 1996]. Annual or seasonal/periodical direct and background emission factors are key parameters for estimating regional N_2O emissions from N-fertilized cropland. The former is determined as the loss rate of fertilizer N applied in the current season/period or year via N_2O emissions, and the latter is calculated the cumulative emissions from a unit area of land free from the application of nitrogenous fertilizers in the current season/period or year. In this study, we calculated the seasonal/periodical or annual direct emission factor (EF_d, in %) of the applied nitrogenous fertilizers (N, in kg N ha^{-1}) using the cumulative emissions from the fertilized treatment (E_F, in kg N ha^{-1}) and the unfertilized field plots (E_0, in kg N ha^{-1}), specifically $EF_d = (E_F - E_0)/N \times 100\%$. The standard error ($SE_{EF}$, in %) of an EF_d was estimated using the standard errors of the three spatial replicates for the E_F (SE_F, in kg N ha^{-1}) and E_0 (SE_0, in kg N ha^{-1}), following $SE_{EF} = 100/N \times (SE_F^2 + SE_0^2)^{1/2}$.

[15] All of the simultaneously measured data on NO emissions included in this report were directly cited from the publication of *Mei et al.* [2009].

[16] We used SYSTAT 5.0 software for Windows (SPSS, Inc., USA) for statistical analyses. The significance levels

for linear and nonlinear regression curves were determined with an *F* test. In addition, we used Origin 8.0 (Origin Lab Corporation, USA) for graph preparation.

3. Results

3.1. Environmental Conditions and Content of Soil Ammonium, Nitrate, and DOC

[17] In addition to the description provided by *Mei et al.* [2009] of the meteorological conditions and soil climate for P1 through P13, we measured the daily precipitation, daily air temperature, soil temperature and soil moisture for P14 in this study. The monthly mean soil temperatures during the four full-year periods ranged from 1.8°C to 3.3°C in January and from 25.8°C to 29.0°C in July, with means of soil temperatures during individual vegetable growing periods varying from 6.9°C to 28.3°C (Table 2). During the entire investigation period, the annual mean air temperature ranged from 15.2 to 16.3°C, with a mean of 15.6°C and a standard deviation (SD) of 0.5°C (Table 3). The annual precipitation varied from 734 to 1,007 mm (mean: 872 mm; SD: 113 mm), with 44–56% of precipitation occurring from June through August and the remaining being distributed more or less evenly across other times of the year. Rainfall, in association with watering, resulted in 753 to 1,026 (mean: 889; SD: 112) mm yr^{-1} of water being input into the vegetable field (Table 3). The soil moisture content during the entire investigation period ranged from 31 to 93% WFPS, with the means for individual periods ranging from 54 to 66% (Table 2).

[18] The levels of NH_4^+-N, NO_3^--N and DOC in the soil are shown in Figure 1. The ammonium levels during the entire investigated period ranged from 2 to 180 (mean: 14) and 0.1–21 (mean: 5) mg N kg^{-1} dry soil (d.s.) in the N-fertilized and unfertilized plots, respectively, with the former being higher than the latter by a factor of 2.9 on average (p < 0.0001). The nitrate levels simultaneously measured in the plots with N fertilizer addition ranged from 0.3 to 212 (mean: 23) mg N kg^{-1} d.s., and these levels were higher than those of the unfertilized plots (ranging from 0.1 to 134 (mean: 9) mg N kg^{-1} d.s.) by a factor 2.5 on average (p < 0.0001). The DOC levels measured from mid-2005 to mid-2006 were 51–226 (mean: 86) and 43–175 (mean: 71) mg C kg^{-1} d.s. in the N-fertilized and unfertilized plots, respectively, with the former being clearly higher, by 21% on average (p < 0.05).

3.2. Nitrous Oxide Fluxes

[19] In the 4 year study period, we conducted a total of 4158 flux measurements and obtained 4063 valid N_2O flux values that were determined to have statistically significant (p < 0.05) initial increase rates of N_2O concentrations against enclosure time. Among these valid fluxes, 71% were above the detection limits for the chamber heights of 0.5 and 1.0 m, respectively. The mean N_2O fluxes of the three replicate plots for both the N-fertilized and unfertilized treatments during the entire investigation period are illustrated in Figure 2. Soon after the application of N fertilizers in nonwinter seasons, the N_2O fluxes from the fertilized plots quickly increased to a maximum and then gradually declined to the prefertilization levels (Figure 2). This occurred following almost all basal fertilization events that
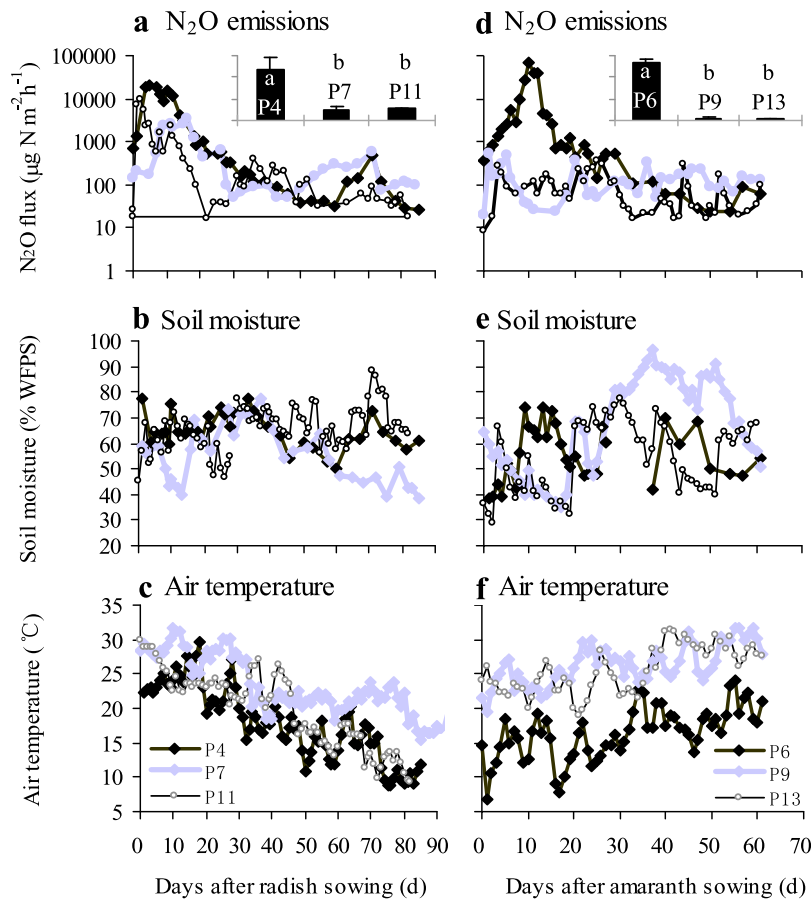
Figure 4. Nitrous oxide (N$_2$O) fluxes during the growing seasons with cultivation of the same vegetable crop in different years and the simultaneously observed soil moisture in the water-filled pore space (WFPS) and air temperature. The given fluxes and soil moisture are the means of three replicate fields, and standard errors are not shown for the purpose of figure clarity. The given temperatures are the means of hourly measured values, and error bars are not shown. (a, b, and c) Radish seasons (P4, P7, P11), and (d, e, and f) amaranth seasons (P6, P9, P13). The inserts in Figures 4a and 4d present the seasonal total N$_2$O emissions in kg N ha^{-1} (see Table 2 for the specific figures). The different letters above the bars of each insert indicate significant differences at p < 0.05.

coincided with soil temperatures ranging from 9 to 29°C (mean: 21°C) [*Mei et al.*, 2009]. However, when soil temperatures were at low levels, N$_2$O emission following N fertilizer addition was not obvious, as was seen for the cases of urea top dressing on 28 February 2005, 27 November 2005, 16 January 2006, 4 March 2007, and 11 March 2008 (Figure 2), when soil temperatures ranged from 3.3 to 12.2°C (mean: 7.2°C) [*Mei et al.*, 2009]. The N$_2$O fluxes from the fertilized plots during the entire investigation period ranged from −36.1 to 66558.7 (mean: 671.2) μg N m^{-2} h^{-1} and were, on average, 28 times higher (p < 0.001) than those from the unfertilized plots (ranging from −2.9 to 391.0 μg N m^{-2} h^{-1}, with a mean of 22.8 μg N m^{-2} h^{-1}).

[20] Of the 14 (P1–P14) experimental periods, five were used for radish crop planting, two for vegetable rape, one for chili, three for amaranth, and two for garlic, and during the remaining short period, the soil was fallow (Table 1). The N$_2$O fluxes for the radish, vegetable rape, chili, amaranth, and garlic growing periods were, on average, 1583, 48, 173,

1532, and 120 μg N m^{-2} h^{-1}, respectively, in the fertilized plots and 22, 28, 16, 20, and 9 μg N m^{-2} h^{-1}, respectively, in the unfertilized plots (Figure 2). As these data show, the highest seasonal/periodical means of N$_2$O fluxes from the fertilized plots appeared in the radish and amaranth growing periods, while the lowest values of the fertilized fields were observed in the vegetable rape seasons.

3.3. Contributions of Peak Emissions to Seasonally/Periodically Cumulative N$_2$O Releases

[21] In the fertilized plots, intensive peak emissions overwhelmingly determined the seasonal/periodical or annual total amounts of N$_2$O released, although they usually occurred within relatively small time spans. In the radish growing seasons, for example, peak emissions occurred during approximately 35% of the seasonal length but accounted for more than 86% of the seasonal cumulative N$_2$O releases. A significant contribution of peak emissions to seasonal cumulative releases also occurred during the rape, chili, amaranth and

a Fertilized

$Log[(NO+N_2O)-N] = 0.32+0.68Log[(NH_4^++NO_3^-)-N]$
$(n=141, R^2=0.13, p<0.001)$

b Unfertilized

$Log[(NO+N_2O)-N] = -0.02+0.59Log[(NH_4^++NO_3^-)-N]$
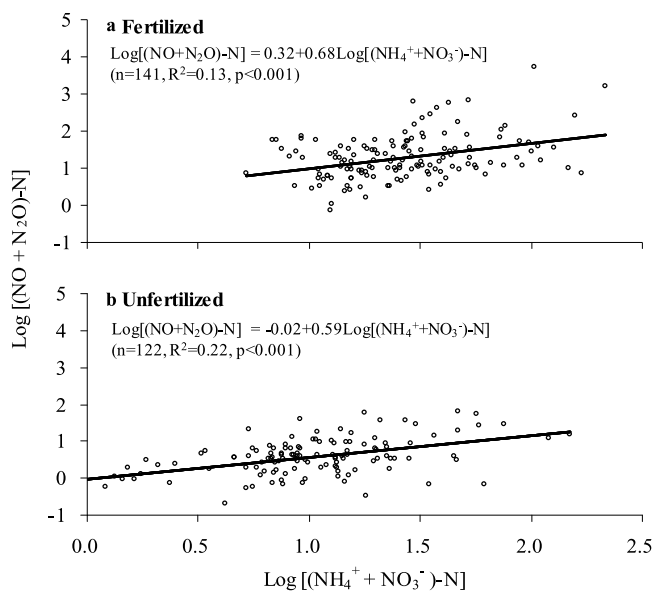$(n=122, R^2=0.22, p<0.001)$

Figure 5. Relationship between the logarithm of the sum of nitric oxide (NO) and nitrous oxide (N₂O) fluxes (in g N ha⁻¹ d⁻¹) and the logarithm of ammonium N plus nitrate N levels in the cultivated soil layer (in mg N kg⁻¹ dry soil).

garlic growing periods. During the entire investigation period, peak emissions from the fertilized plots contributed over 86% of the total amount of N₂O emissions.

3.4. Effects of Temperature and N Addition on Seasonal/Periodical Total N₂O Emissions

[22] The cumulative N₂O emissions for the 14 periods of both field treatments are listed in Table 2. The cumulative N₂O emissions from the fertilized field plots in individual full vegetable seasons (P1, P10 and P14 were excluded) varied from 0.56 (in P5) to 54.7 (in P6) kg N ha⁻¹, with a CV of 153% for the 11 observations. The total N₂O emissions in the 11 individual full vegetable seasons of the unfertilized plots ranged from 0.15 (P13) to 1.48 (P2) kg N ha⁻¹, with a CV of 88%.

[23] When we plotted the seasonally/periodically accumulated N₂O emissions (N₂O_ST, in kg N ha⁻¹) from both the fertilized and unfertilized fields against the product of the corresponding seasonal/periodical averages of soil temperatures (5 cm depth; T_sa, in °C) and quantities of applied N fertilizers (F_N, in t N ha⁻¹), an exponential correlation clearly appeared (Figure 3a), even though significant deviation occurred for the case of P6, which showed an extremely high cumulative emissions level. The correlation could be described with an exponential equation (see the regression curve in Figure 3a). As the determination coefficient (R^2) of this nonlinear regression indicates, interactions between soil temperatures and quantities of applied nitrogen fertilizers could explain up to 73% of the variation in the seasonally/periodically accumulated N₂O emissions. These results suggest that the seasonal/periodical variations in N₂O emissions from the conventional vegetable fields were mainly regulated jointly by soil temperature and nitrogen availability. The equation shown in Figure 3a implies

that direct extrapolation of any field measurements made during a short period of less than a year to the annual or subdecadal scale will most likely yield considerable uncertainties.

3.5. Direct and Background N₂O Emissions Factors

[24] Based on the seasonally/periodically accumulated quantities of N₂O emissions from the fertilized and unfertilized treatments and the N fertilizer addition rates (Table 2), we estimated the EF_ds for individual vegetable growing periods. As shown in Table 2, the EF_ds of individual full vegetable seasons ranged from 0.06 to 14.2%, with a mean of 3.09% (data from P1, P10 and P14 were excluded). These seasonal EF_ds showed high variation, with a CV of up to 142%. Further analysis indicated that approximately 43% of the variation in the EF_ds could be explained by the interaction of soil temperature and the rates of applied N fertilizers. This is illustrated by the regression equation presented in Figure 3b.

[25] The equation shown in Figure 3b suggests that, in a vegetable field, the conversion rates of applied N fertilizers to N₂O are stimulated by the interaction of soil temperature and N availability. This clearly indicates that an EF_d determined from field observations during a short period will underestimate or overestimate N₂O emissions if it is directly extrapolated to annual and subdecadal scales while ignoring the seasonality in soil temperature, interannual variations in climate and seasonal/periodical or interannual unevenness of fertilizer distribution.

[26] Using the year-round observations from the four study years listed in Table 2, the annual total emissions of the fertilized and unfertilized plots were estimated and are presented in Table 3. The N₂O annual emissions amounted to 8.2–66.6 and 0.51–4.14 kg N ha⁻¹ yr⁻¹ in the fertilized and unfertilized treatments, respectively. The annual EF_ds were determined to be 2.88 ± 2.14% (standard deviation of four observations) on average. This mean value is larger than the *IPCC* [2006] default (1.0%) by a factor of approximately 3, and implies a substantial contribution of vegetable cultivation to regional N₂O emissions from croplands. The annual cumulative release from the unfertilized plots provided annual background N₂O emissions factors of 1.81 ± 1.73 (standard deviation of 12 observations: three spatial replicates, each with four interannual replicates) kg N ha⁻¹ yr⁻¹, which is equivalent to approximately 5% of the direct emissions. Interannual CVs of 74 and 96% were found for the annual direct and background emission factors, respectively. These high interannual CVs highlight the necessity of conducting multiple-year field measurements for quantifying emission factors.

3.6. Variation in N₂O Emissions Among Nonwinter Seasons

[27] Huge variations in seasonal EF_ds also occurred for nonwinter vegetable seasons, not only among different vegetable crops, but also for the same vegetable species cultivated at comparable times in different years. For instance, the CVs of the EF_ds were 76% for the three full seasons of radishes cultivated in 2005 (P4), 2006 (P7) and 2007 (P11) and 137% for the three full seasons of amaranth planted in 2006 (P6), 2007 (P9) and 2008 (P13). The total N₂O emissions in P4 and P6 were significantly higher, by factors
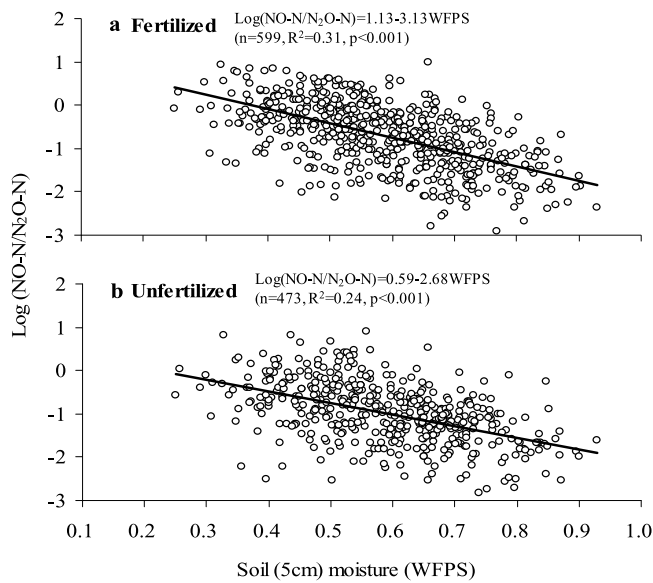
Figure 6. Relationship between the logarithm of the ratios of nitric oxide (NO) to nitrous oxide (N$_2$O) fluxes (in g N ha^{-1} d^{-1} for either gas) and soil moisture in the water-filled pore space (WFPS). The fluxes of N$_2$O originated from this study, and the simultaneously measured NO fluxes and soil moisture were adapted from *Mei et al.* [2009].

of 4–5 (p < 0.05) and 25–35 (p < 0.001), respectively, compared with the other two growing seasons for the same vegetable species (Figures 4a and 4d). As a result, the EF$_d$s in P4 and P6 were 2- to 18-fold higher than for the other full seasons of the same vegetable crops (Table 2).

3.7. Relationship Between NO Plus N$_2$O Emissions and Mineral Nitrogen Contents

[28] According to the "hole-in-the-pipe" conceptual model, the amount of NO plus N$_2$O emitted from ecosystems is a function of the extractable inorganic nitrogen. Our data show a significant linear correlation between the logarithms of NO plus N$_2$O mass fluxes (in g N ha^{-1} d^{-1}) and the logarithms of extractable inorganic nitrogen levels (in mg N kg^{-1} d.s.), with slopes varying from 0.59 ± 0.10 (standard deviation; same below) for the unfertilized plots to 0.68 ± 0.15 for the fertilized fields and intercepts from −0.02 ± 0.11 to 0.32 ± 21, respectively (Figure 5). The parameters were similar those reported by *Davidson and Verchot* [2000] for pasture, forest, and agricultural lands (with slopes ranging from 0.53 to 0.79 and intercepts from −0.24 to 1.08).

3.8. Relationship Between NO to N$_2$O Ratios and Soil Moisture

[29] Using the N$_2$O fluxes shown in Figure 2 and the simultaneously measured NO fluxes cited from *Mei et al.* [2009], we calculated the logarithms of the NO to N$_2$O ratios of the daily released nitrogen mass on a hectare basis (g N ha^{-1} d^{-1}). Negatively linear relationships significantly (p < 0.001) appeared in both the fertilized and the unfertilized plots (Figure 6) between the logarithms of the ratios and the simultaneously observed soil moisture (at 0–5 cm

depth) in the water-filled pore space (WFPS), which was expressed as fractions of 0–1 (adapted from *Mei et al.* [2009]).

4. Discussion

4.1. Uncertainties in Estimates of Annual or Subdecadal Emissions

4.1.1. Uncertainties Associated With Temporal Resolution of Measurements

[30] Almost all available data on N$_2$O emission fluxes from terrestrial ecosystems originate from intermittent measurements made using static chamber techniques [e.g., *Stehfest and Bouwman*, 2006] (www.mnp.nl/en/publications/2006). Scientists are often concerned with the uncertainties induced by intermittent field measurements for seasonal/periodical or annual estimates, which usually poorly represent the intrinsic high temporal variability in N$_2$O emission from terrestrial ecosystems [e.g., *Lin et al.*, 2010]. Taking into account the 12–13% overestimation of daily emissions under clear weather by single measurements made at 09:00–11:00 LST [*Yao et al.*, 2009; *Liu et al.*, 2010], the annual occurrence frequency of 40% for clear weather days during our investigation period, and that in gap fillings of missing daily fluxes the positive and negative errors were almost offset by each other, direct extrapolation of our daily single measurements could overestimate the annual cumulative emissions by only about 4%. As the latest study [*Liu et al.*, 2010] shows, this error could be even avoided if the daily single measurements were performed one hour earlier than the time of our study.

4.1.2. Uncertainties Associated With Spatial Replicate Number of Measurements

[31] As the results stated above show, three spatially replicated measurements for each field treatment still poorly covered the large spatial variability, and then led to large uncertainties in the estimates of seasonal/periodical and annual cumulative emissions (with spatial CVs of 9–105% (mean: 40%) and 4–82% (mean: 28%), respectively). This implies that increasing the number of spatial replicates should be a priority for reducing the current uncertainties in our estimates of annual total emissions, as well as EF$_d$s.

4.1.3. Uncertainties Induced by Short-Time Observations or a Lack of Interannual Replicates

[32] Our results indicate that short-period measurements may result in very large uncertainties in the estimation of annual or subdecadal N$_2$O emissions from open vegetable fields. Using the observations listed in Tables 1 and 2, we simulated the magnitude of uncertainty resulting from simple extrapolation of short-term measurements. With regard to EF$_d$s, simply extrapolating the observations of a particular vegetable season to an annual scale could produce uncertainty magnitudes of 4% to 23-fold (with a mean of 120%); using the observations of a certain year to represent the EF$_d$ of another year could yield an overestimation or underestimation of 10% to sevenfold (with a mean of 90%), while it could underestimate or overestimate subdecadal direct emission factors by 50–70%. These magnitude figures indicate that applying the emission factor approach recommended by the *IPCC* [2006] Tier 2 guidelines, but using observed emission values that poorly represent seasonality and/or interannual variability will inevitably yield large uncertainties in the

estimates of annual N$_2$O emissions from vegetable fields for those years lacking measurements. Apparently, it is necessary to identify the factors regulating N$_2$O emissions from vegetable fields and to understand their functional mechanisms to develop better estimation approaches, such as empirical or process-oriented models.

4.2. Factors Regulating N$_2$O Emissions

4.2.1. Temperature-Determined Low Direct Emission Factors in Winter Seasons

[33] The smallest EF$_d$s of full individual vegetable seasons were found whenever winter vegetables (e.g., vegetable rape and garlic) were cultivated (Table 2). Our results suggest that the seasonal EF$_d$s became larger either the earlier a winter vegetable season was started or the later it was terminated. With regard to vegetable rape, for instance, the EF$_d$s decreased from 0.18% for the season terminated in late May to 0.06% for the one that ended in late March, though the N fertilizer addition rate of the latter period was 30% higher. The mean topsoil temperature was 8°C on average in the four winter vegetable seasons, with a mean seasonal EF$_d$ of 0.34%, but it was 23°C on average in the nonwinter vegetable seasons, with a mean EF$_d$ of 1.81% (P4 and P6, which showed extremely intensive emissions, were excluded from these statistics). The significantly different temperature (p < 0.001) very likely accounted for the several-fold difference in the EF$_d$s between the winter and nonwinter vegetable seasons (p < 0.01). This implies that applying more inorganic and organic fertilizers to winter crops could be a mitigation option for N$_2$O emissions, as the added nitrogen and carbon can be immobilized by microbes instead of being immediately denitrified to N gases and then slowly released later for nonwinter crops.

4.2.2. Moisture- and Substrate-Dominated N$_2$O Emissions in Nonwinter Seasons

[34] The much higher EF$_d$s observed in P4 and P6 than in others (Table 2) were most likely due to: (1) a greater amount of nitrogen substrate being available from the basal fertilizers for nitrification and denitrification, whereby N$_2$O is produced as a byproduct or an immediate product; (2) more favorable soil moisture conditions for N$_2$O production by denitrification immediately following fertilizer incorporation; or (3) a greater amount of carbon substrate being available from the application of lagoon-stored organic slurry. The first explanation may be supported by the data on nitrogen addition rates listed in Table 2. While the same fertilizers (urea plus soybean cake) were basally applied in the three radish seasons (Table 1), the addition rate of pure N in P4 was 31–38% higher than in P7 and P11. Similarly, the fertilizer N basally incorporated in P6 was higher by factors of 1.9 and 3.2 compared with P9 and P13, respectively. The higher N addition rates might have provided more N substrate for the microbial processes related to N$_2$O production and, thus, stimulated more intensive emissions. The second reason may be supported by the precipitation and soil moisture data [*Mei et al.*, 2009]. Of the seasonal total precipitation, 84% fell in the early stage (from three days before to 20 days after incorporation of basal fertilizers) of P4 but only 41% fell in the same stage of P7 or P11. The higher precipitation led to significantly higher (p < 0.05) soil moisture in the early stage of P4 than in the same stage of the other two radish seasons (85% versus 74%

WFPS on average) (Figure 4b) and obviously lower (P < 0.05) air temperatures by 1.8–5.1°C (Figure 4c). Of the seasonal total precipitation in P6, P9, and P13, 29%, 10%, and 25% fell in the early stage of the respective amaranth seasons [*Mei et al.*, 2009]. Although the precipitation in the early stage of P6 totaled only 46 mm, a single intensive rainfall event of 33 mm occurring on the eighth day after fertilizer incorporation led to a rapid increase in soil moisture to a level of approximately 82% WFPS. This soil moisture was higher by 24% on average (p < 0.001) than in P9 and P13 and remained until the end of the early stage (Figure 4e), which was most likely due to the much cooler air temperature in P6 (lower by 10–12°C on average; p < 0.001) because this season started in spring, approximately two months earlier than the other amaranth seasons. The significantly higher soil moisture might have facilitated N$_2$O production through microbial denitrification and, thus, stimulated more intensive emissions. The third reason is likely to particularly account for the extremely high EF$_d$ observed in P6. In this season, organic fertilizers in the form of human manure slurry from a locally typical indoor lagoon and soybean cake were incorporated into the soils prior to the sowing of amaranth seeds. This was different from what took place in P9 and P13, in which a mineral fertilizer and organic manure in the form of rapeseed cake were basally applied. Before the human manure slurry was applied to the fields, it was stored for at least several months in an indoor lagoon, where ammonium from the hydrolysis of the urea in urine and the mineralization of organic excreta might have been well maintained in a water-saturated environment, and DOC from the decomposition of organic matter might also have accumulated. The manure slurry removed from the indoor lagoon was further diluted with water before it was applied. When the water-saturated manure slurry was basally applied, intensive denitrification might have occurred in the bare soils and could have resulted in intensive production of N$_2$O. Our simultaneous measurements of nitrate and DOC levels (Figures 1b and 1c) showed DOC-to-nitrate molar ratios of approximately four in P4 and P6. These ratios indicate that there was no limitation of the available carbon substrates upon denitrification, provided that this microbial process required 5 mol of DOC to reduce 4 mol of nitrate [*Swerts et al.*, 1996; *Ingwersen et al.*, 1999]. The application of lagoon-stored organic slurry in the amaranth season might have resulted in more intensive denitrification in the early period and, thus, induced a much higher seasonal fertilizer N-loss rate via N$_2$O emissions compared with the radish season (Table 2), even though soybean cake was basally applied in both P4 and P6; the DOC-to-nitrate molar ratios and soil moistures were similar during the time with the most intensive N$_2$O emissions; and the early temperature was much lower in P6 than P4 (Figure 4). The stimulatory effect of lagoon-originated manure slurry on N$_2$O emissions can probably be attributed to three causes. First, the population of denitrifiers was probably increased by slurry application, as during storage, the organic manure with saturated water content, rich available nitrogen and labile carbon presented favorable conditions for the growth of anaerobic microbes [e.g., *Swerts et al.*, 1996]. Second, the N$_2$O:N$_2$ ratios of denitrification products were likely increased because this process occurred in aerobic soil during incomplete anaerobic conditions that were favorable for immediate production [e.g., *Poth and Focht*, 1985]. Ultimately, nitrification

was likely stimulated by the addition of ammonium-rich manure slurry, which produced N$_2$O by itself while further stimulating N$_2$O production through subsequent denitrification [e.g., *Williams et al.*, 1992]. In the present study P6 was the only case in which lagoon-stored manure slurry was added. This most likely accounted for the fact that both the seasonal total amount and direct factor of N$_2$O emissions in this season greatly deviated from the regression curves illustrated in Figure 3. The above results likely indicate "hot spot" agricultural sources of N$_2$O emissions in circumstances in which lagoon-stored manure slurry is applied. However, further study is required to address this issue because it has been very poorly investigated to date.

4.2.3. Mitigation Options for N$_2$O Emissions

[35] Regarding the typical and conventional vegetable fields investigated in this study, the discussion above shows that temperature, supplies of nitrogen and carbon substrates, soil moisture and microbial activities appeared to be the key factors that regulated not only the total amounts, but also the direct factors related to seasonal N$_2$O emissions. Whenever nitrogen availability, labile carbon supplies, soil moisture and the population of denitrifiers were simultaneously at high levels in nonwinter seasons, explosive N$_2$O emissions from the conventional vegetable fields could alone induce fertilizer nitrogen losses at rates of a few percent to much more than ten percent. Otherwise, temperature became the overwhelming inhibitor in winter, while the quantity of available nitrogen became the dominant regulator in nonwinter seasons. Based on our results, certain practices are expected to decrease denitrification and, thus, to mitigate N$_2$O emissions from conventional open vegetable fields. These are (1) reducing basal N addition rates while only using organic slurry in dry and cool seasons, (2) splitting N addition and watering and avoiding N application before heavy rainfall or irrigation, and (3) splitting single or double applications of N fertilizers into multiple applications and distributing more fertilizer N in periods with intensive vegetable plant uptake. However, the mitigation effects of these measures still require further testing in future experiments.

4.3. Effects of Mineral Nitrogen on NO Plus N$_2$O Emissions

[36] The slopes and intercepts of the linear regression for the logarithms of NO plus N$_2$O mass fluxes against the logarithms of extractable inorganic nitrogen levels (Figure 5) were within the ranges observed for other terrestrial ecosystems excluding rice paddy-based croplands [*Davidson and Verchot*, 2000]. Compared with the nonrice period of a fertilized rice-wheat rotation ecosystem located at another field site with a silt clay soil in the same delta region [adapted from *Yao et al.*, 2010], the slope observed in our fertilized vegetable plots was larger (0.68 versus 0.41 on average), but the intercept was smaller (0.32 versus 1.36 on average). However, both parameters of our unfertilized vegetable plots were very similar to those (adapted from *Yao et al.* [2010]) observed in the nonrice period of a fertilized rice-wheat rotation field with the same sandy loam soil that was located beside our experimental fields (0.59 versus 0.69 on average for slopes and −0.02 versus −0.03 on average for intercepts). These results indicate that large differences in soil properties may lead to different parameter values for the "hole-in-the-

pipe" model. However, the factors determining the parameters remain unclear. Further investigation is still needed to address this issue.

4.4. Regulatory Effects of Soil Moisture on NO to N$_2$O Ratios

[37] The significant relationships shown in Figure 6 logically indicate that increasing amounts of N$_2$O emissions could be attributed to microbial denitrification with increasing soil moisture. Though the slopes (−3.13 to −2.68) and intercepts (1.13 to 0.59) of the linear regressions varied between the two fertilization treatments, the ranges of both parameters were much narrower compared with those found for the nonrice (nonwaterlogged) periods of different rice-wheat rotation field sites in the Yangtze River delta (adapted from *Yao et al.* [2010]), which exhibited slopes of −6.11 to −1.84 (mean: −3.17) and intercepts of 0.84 to 4.70 (mean: 1.93). In comparison with the values of these parameters determined for global major terrestrial ecosystems excluding paddy rice fields (slope: −6.14 to −0.93, with a mean of −3.97; intercept: 0.64 to 3.82, with a mean of 2.73) reported by *Davidson and Verchot* [2000], those of our vegetable field and the nonwaterlogged rice-wheat fields mentioned above showed similar CVs (51% versus 79% for slopes and 98% versus 70% for intercepts). The relationships between the logarithms of the NO to N$_2$O ratios and soil moisture have ever been assumed to be applicable for global inventory studies on the emissions of these two gases from terrestrial ecosystems [*Davidson and Verchot*, 2000]. However, the parameters measured in this study, as well as those reported in the literature, appear to be specific for ecosystem types, soil properties, or management practices. In fact, factors determining the parameters have not been well investigated. This situation continues to limit the applicability of such empirical models to global or regional inventory studies. Understandably, further investigation is needed to identify key factors determining the relationships between the NO to N$_2$O ratios and soil moisture.

5. Conclusions

[38] Nitrous oxide (N$_2$O) emission from typical vegetable fields subject to conventional management practices were measured over a 4 year period spanning 1,475 days. Peak releases following nitrogen addition overwhelmingly accounted for the annual cumulative N$_2$O emissions and the total emissions of individual vegetable growing seasons. Temperature, availability of nitrogen and carbon substrates, soil moisture and microbial activity stimulated by organic slurry application appeared to be the key factors that regulated not only the total amounts of but also the direct factors related to seasonal N$_2$O emissions. Whenever these factors were simultaneously at high levels, explosive N$_2$O emissions, alone, could induce fertilizer nitrogen losses in individual vegetable seasons at rates of a few percent to much more than ten percent. Otherwise, temperature was the overwhelming inhibitory factor in winter-crop seasons, while the quantity of available nitrogen became the dominant regulator in nonwinter seasons. Direct N$_2$O emissions factors varied from 0.06 to 14.2% among individual crop-growing seasons and varied annually from 0.59 to 4.98%, with a mean of 2.88% (and an interannual CV of 74%), which was much higher than

the default value of the IPCC guidelines. Using emission factors obtained from short-term measurements is expected to yield large uncertainties in the inventory of N$_2$O emissions from open vegetable fields. To reduce the current uncertainties in annual direct emissions factors, increasing the number of spatial replicates in multiple-year measurements is recommended as a priority when the temporal resolution used this study is adopted. The results of this study suggest some potential practices for decreasing denitrification and, thus, mitigating N$_2$O emissions from conventional open vegetable fields, which are (1) reducing basal N addition rates, while only using organic slurry in dry and cool seasons, (2) splitting N addition and watering and avoiding N application before heavy rainfall or irrigation, and (3) splitting single or double applications of N fertilizers into multiple applications, while distributing more fertilizer N in winter crop seasons or in nonwinter periods with intensive vegetable plant uptake. However, the mitigation effects of these options still require further testing in future experiments.

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J. Deng, H. Dong, C. Liu, R. Wang, B. Xie, Z. Yao, X. Zheng, and Z. Zhou, State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China. (xunhua.zheng@post.iap.ac.cn)

B. Mei, College of Environment and Resources, Inner Mongolia University, Huhhot 010021, China.

J. Zhu, State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China.