

Nitrous Oxide Emissions from Household Vegetable Fields in the Rural Residential Areas of Hilly Subtropical Central China

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Abstract

Nitrous oxide (N_2O) emissions from a household vegetable field in a rural residential area of hilly subtropical central China were observed using a static chamber-gas chromatographic method from January 2010 to December 2011. The N_2O fluxes exhibited seasonal dynamics and the accumulated N_2O emissions during the wet seasons accounted for 83.5% of the total N_2O emissions. Soil mineral nitrogen (N) contents were found not limiting factors because of the application of large amounts of human excreta. The daily N_2O fluxes showed a significant, positive correlation with soil temperature, soil moisture and soil NO_3^- -N content, and soil denitrification may be the major pathway responsible for N_2O emissions. High-frequency, intensive application of liquid excreta stimulated the N_2O emission process. The average annual N_2O emission rate was 12.1 ± 0.9 kg N ha⁻¹ year⁻¹ in the examined household vegetable field, and the total N_2O emissions from household vegetable fields originating from the N source of human excreta in the studied Jinjing catchment (135 km²) were estimated as 1.58 ± 0.16 ton N year⁻¹. Such emissions can be considered as N_2O re-emissions of the N input into the ecosystem, and the emission factor of N_2O re-emissions was estimated to be 0.57%. The findings indicated that under the present management practices, household vegetable fields in the subtropics of China provide a relevant contribution to greenhouse gas emissions and a responsive mitigation scheme at a household scale is needed to reduce N_2O emissions.

Keywords: N_2O ; Household vegetable field; Rural residential area; Subtropics

Abbreviations: MCMC: Markov Chain Monte Carlo; PCA: Principal Component Analysis

Introduction

Nitrous oxide (N_2O), an important greenhouse gas in the atmosphere, displays high global warming potential that is 298 times greater than that of carbon dioxide (CO_2) on a 100 year time scale [1]. With a steady-state lifetime of approximately 120 year, N_2O plays an important role in ozone depletion in the stratosphere [2]. Mosier and Kroeze [3] reported that the atmospheric N_2O concentration will reach 340-350 ppbv in 2020 resulting from food production systems and Montzka et al. [4] found that the atmospheric N_2O concentration was already 322 ppbv in 2011 and increased at a mean rate of 0.7 ppbv year⁻¹ during the past 30 years. Agricultural soil, which exhibited a total N_2O flux of 6.2 Tg N_2O -N year⁻¹ and accounted for 35% of the total global annual N_2O -N emissions, has been considered as a major source of global warming [5]. Soil N_2O emissions primarily result from two important microbial activities, nitrification and denitrification, and are directly influenced by factors related to nitrogen (N) fertilizers, such as fertilization quantity, fertilizer type and timing of fertilization [3].

Many intensive vegetable production systems were not sustainable because of excessive N fertilizer application, resulting in soil acidification, ammonia volatilization, nitrate leaching, soil erosion, nitrification and denitrification [6]. Vegetable fields, with a total cultivation area of 20.35 million ha, account for 12.45% of the total sowing area of crops in 2012 in China [7]. More importantly, intensive vegetable production systems were estimated to constitute approximately 17% of the total N consumption by agricultural systems in China [8]. Some intensive vegetable fields were characterized by very high N application; for example, the N application rate in vegetable fields was 1,340 kg N ha⁻¹ year⁻¹ in Jiangsu Province and 1,710 kg N ha⁻¹ year⁻¹ in Shandong

Province [9]. An extremely high N application rate of 3,714 kg N ha⁻¹ year⁻¹ was reported in a rotation vegetable field in suburban Nanjing, Jiangsu Province [10]. However, numerous studies have demonstrated that excessive N fertilizer application can promote N_2O emissions from soils [8] and that the emission factor increases when the N application rate exceeds the plant N requirements [11]. In some cases, N_2O emissions from intensive vegetable fields were significantly affected by the specific rotation pattern [10]. Many investigations have been performed to evaluate N_2O emissions from vegetable fields in different countries, such as the Netherlands [12], Niger [13], the United States of America [14], Australia [15], Germany [16], Japan [17] and China [9,10,18,19] and all of these studies revealed the high environmental risk of greenhouse gas emissions from vegetable fields. For example, a 6 year observation of onion cultivation systems in Japan revealed that the N_2O emissions ranged from 3.5 to 15.6 kg N ha⁻¹ specifically during the growing season (from April to October) [17]. Compared to cropland and forest soils, vegetable fields had the greatest N_2O emission rate because these fields had the greatest N application rate [19]. Moreover, Jia et al. [10] reported that the annual N_2O emissions from a rotation vegetable field to which an extremely high amount of N fertilizer was applied ranged from 56.4 to 238 kg N ha⁻¹ year⁻¹. Because of increasing attention on the high risk of greenhouse gas emissions from vegetable cultivation systems, many studies have focused on N_2O

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Received October 18, 2016; **Accepted** October 24, 2016; **Published** November 04, 2016

Citation: Fu X, Li Y, Wang Y, Shen J, Xiao R, et al. (2016) Nitrous Oxide Emissions from Household Vegetable Fields in the Rural Residential Areas of Hilly Subtropical Central China. Environ Pollut Climate Change 1: 103. doi: [10.4172/2573-458X.1000103](https://doi.org/10.4172/2573-458X.1000103)

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emissions from large-scale commercial vegetable fields and greenhouse systems [10,12,15,18,20]. To date, few investigations have focused on N₂O emissions from household vegetable fields in rural residential areas, which are cultivated using a very different system in which liquid excreta from nearby septic tanks are used at very high rates as the only N input. N₂O emissions from these fields have been overlooked due to their easily-neglected small size and highly scattered distribution surrounding rural residential houses. Importantly, N is consumed daily by humans through food (grain, meat, milk, etc.) and this N is removed from the ecosystem. For example, plants, including grains, vegetables and fruits, absorb N from the soil and livestock and poultry take up N from grains or grass, resulting in the loss of N from the ecosystem. In the N cycling process, human excreta return to the soil as N fertilizer; thus, the N₂O flux originating from the N in human excreta may be regarded as N₂O re-emission. The few studies that focused on N₂O re-emission in an ecosystem categorized this N₂O as direct emissions rather than re-emissions [21]. Thus, N₂O emissions from this unique land use type in rural residential areas may warrant further exploration to determine its contribution to the regional greenhouse gas emission inventory.

Therefore, we chose a typical agricultural catchment in a hilly red-soil region of subtropical central China to survey the spatial characteristics of household vegetable fields surrounding rural residential areas. We measured the N₂O emissions from these small-size and sparsely distributed household vegetable fields with the following ultimate aims: i) to analyze the seasonal dynamics of N₂O emissions and the environmental factors affecting these dynamics; ii) to estimate the annual N₂O emissions and the direct N₂O emission factor specific to household vegetable fields; and iii) to evaluate the contribution of these fields to N₂O emissions to diminish the uncertainty concerning the regional inventory of greenhouse gas emissions.

Materials and Methods

Site description

The field experiment was carried out at the Changsha Research Station for Agricultural and Environmental Monitoring, Institute of Subtropical Agriculture, Chinese Academy of Sciences, Changsha, Hunan Province, China (113°20'E, 28°35'N) (Figure 1). This region has a subtropical monsoon climate, with a mean annual air temperature of 17.5°C (1979-2010) and a mean annual precipitation of 1,330 mm (1979-2010). The soil type is Haplic Alisols, which developed from the parent material of granite and are widely distributed throughout Hunan Province. The soil had a sandy loam texture and the clay, sand and silt contents were 24.7, 36.6 and 38.7%, respectively. The soil was slightly acidic, with a pH of 5.7, and the soil bulk density was 1.35 g cm⁻³. The contents of soil organic carbon (C), total soil N and total soil phosphorous in the topsoil (0-20 cm) were 14.2, 2.5 and 1.27 g kg⁻¹ dry soil, respectively.

Vegetables are produced to nearly self-sufficiency in rural areas; local residents plant their own vegetable fields near their houses to satisfy the families' need for vegetables rather than purchasing them from the market. Each family owns a plot that is small in size and that has predominantly been transformed from woodland, especially Masson pine forest, for vegetable cultivation. A household vegetable field (300 m²) in a rural residential area was chosen for N₂O emission observation and managed and cultivated in a typical and conventional manner according to the planting habits of local residents. The field observation spanned from January 2010 through December 2011. The cultivation was designed with three vegetable rotations throughout

each year: bok choy (*Brassica campestris* L. ssp. *chinensis* Makino), hot pepper (*Capsicum annuum* L.) and purple flowering stalk (*Brassica campestris* L. var. *purpurea* Bailey) in 2010 and greens (*Brassica chinensis* L. var. *chinensis*), jicama (*Pachyrhizus erosus* L.) and radishes (*Raphanus sativus* L.) in 2011 (Table 1). During the observation period, only liquid excreta (human excrement) were applied as N fertilizer. All field management practices were conducted by a local farmer according to local practices. Accordingly, the N application timing and irrigation activities were irregular because of uncertainty concerning weather conditions and the growth needs of the plants. Consequently, accurately calculating the N input was very difficult. Based on results reported by Liu et al. [22] and the fact that there were three persons in a family with 6.19 kg N year⁻¹ input per person, and the total N input was estimated to be approximately 619 kg N ha⁻¹ year⁻¹ in our studied vegetable field.

Nitrous oxide measurement

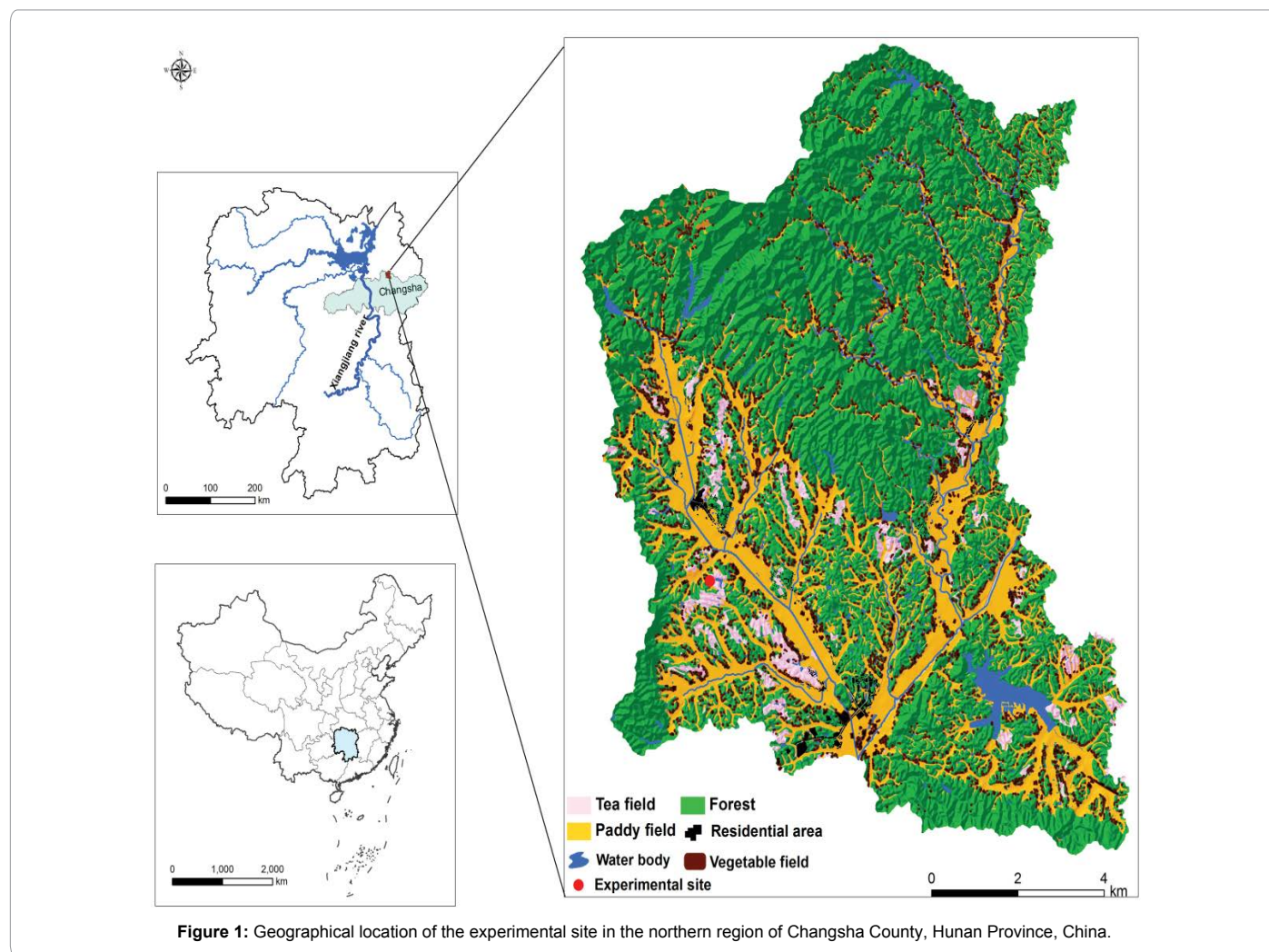
The in situ observation of N₂O emissions was performed using the static closed chamber-gas chromatography (GC) method. The entire chamber system consisted of two components, the chamber and the base; the chamber was 0.5 m tall and the base covered an area of 0.4225 m². The chamber was sealed with clean water as gas samples were collected. Simultaneously, the air inside the chamber was mixed using two small fans (10 cm diameter) that functioned using 12 V batteries. The experiment included three replicates, and the chambers were installed at 7-8 m intervals. Gas sampling was performed at 09:00-11:00 AM every week throughout the observation period. Five gas samples were collected from the headspace of each chamber using a 100 ml syringe equipped with three-way stopcocks at 0, 10, 20, 30 and 40 min following chamber closure; then, the samples were stored in pre-evacuated 12 ml vials fitted with a rubber septum until GC analysis. The temperatures inside the chamber and of 0-5 cm topsoil (T_{soil}) were monitored simultaneously using a JM624 portable digital thermometer (JinMing Instrument Co. Ltd., China). The N₂O concentrations were analyzed within 24 h using a gas chromatograph (Agilent 7890A, Agilent, USA) equipped with a ⁶³Ni-electron capture detector and an automated sample injector system, which was calibrated using commercial cylinders containing standard gases of N₂O in air in the range of 0.3-3 ppmv. Daily N₂O flux was calculated using a linear regression between the N₂O concentrations, and the sampling times; and the annual N₂O emission rate was calculated using a method described by Fu et al. [23].

Soil and environmental variables

Fresh soil samples (0-20 cm) were collected using an auger after gas sampling was complete. The soil samples were collected near the base in the same plots, placed in clean zip-lock bags to avoid soil moisture evaporation, and stored in a refrigerator at 4°C. Before analysis, the soil samples were homogenized manually and sieved through a 2 mm

| Vegetable | Planting time | Fertilizer | Total N application (kg N ha ⁻¹ year ⁻¹) |
|------------------------|---------------|-----------------|---|
| Bok choy | 9 Nov 2009 | Human excrement | |
| Hot pepper | 12 Apr 2010 | Human excrement | |
| Purple flowering stalk | 25 Sep 2010 | Human excrement | |
| | | | 619 |
| Greens | 10 Jan 2011 | Human excrement | |
| Jicama | 2 Apr 2011 | Human excrement | |
| Radishes | 8 Sep 2011 | Human excrement | |
| | | | 619 |

Table 1: Vegetable species and fertilization.



mesh to remove stones, aboveground plant materials and plants roots from the samples. Soil volumetric water content (SWC) was analyzed using the oven-drying method (105°C, 24 h). Soil ammonium-nitrogen (NH_4^+-N) and nitrate-nitrogen (NO_3^--N) content were analyzed using an automated flow injection analyzer (Fiastar 5000). All measurements were replicated four times. Additional methodological details may be found in Fu et al. [23]. Daily minimum temperature (T_{\min} , °C), daily maximum temperature (T_{\max} , °C), daily mean temperature (T_{air}) and daily precipitation (mm d^{-1}) were recorded by a representative weather station (Inteliment A, IMET-ADV2, Dynamax, USA) not far (~200 m) from the chosen household vegetable field in the rural residential area.

Survey and estimation of the total area of household vegetable fields

There was no precise record of the total area of household vegetable fields in the study catchment due to their small size and highly scattered distribution. Therefore, a survey was conducted from 23 April 2013 to 26 April 2013 and 38 families were surveyed to determine characteristics such as the number of family members, the area of household vegetable fields and the housing area (Table 2). Based on these results, it was assumed that the area of household vegetable fields for each person (one person) and each housing area unit (1 m^2) were identical throughout the catchment and that the vegetable field

area of the household displayed a binary linear correlation with the number of family members and the housing area. These are reasonable assumptions for this small catchment displaying similar natural and anthropogenic characteristics (i.e., soil fertility, climate, land management and socioeconomic conditions). Based on the variations in the survey results for the number of family members, the housing area, and the household vegetable field area, we calculated the binary linear regression equations between vegetable field area, number of family members and housing area using the Bayesian Markov Chain Monte Carlo (Bayesian MCMC) analysis tool (OpenBUGS, Cambridge Institute of Public Health, UK). Therefore, the total household vegetable field area in the study catchment was estimated based on a binary linear regression equation considering the total housing area and the population in the catchment. The total housing area in the catchment was obtained from the statistical data of a vectorized aerial map, and the population data were from the statistical data provided by the local government.

The total N_2O emissions in the Jinjing catchment

The total N_2O emissions from household vegetable fields in the Jinjing catchment were estimated as the product of the annual N_2O emissions per unit area and the total household vegetable field area in the study catchment.

| No. | Family members (person) | Household vegetable size (m ²) | Housing area (m ²) | Village name |
|-----|-------------------------|--|--------------------------------|--------------|
| 1 | 5 | 72 | 200 | Xishan |
| 2 | 5 | 54 | 160 | Xishan |
| 3 | 2 | 81 | 340 | Xishan |
| 4 | 5 | 100 | 273 | Xishan |
| 5 | 4 | 154 | 156 | Xishan |
| 6 | 4 | 81 | 200 | Xishan |
| 7 | 6 | 68 | 110 | Xishan |
| 8 | 5 | 54 | 150 | Xishan |
| 9 | 2 | 52 | 150 | Xishan |
| 10 | 2 | 76 | 172 | Xishan |
| 11 | 4 | 113 | 125 | Xishan |
| 12 | 2 | 115 | 184 | Xishan |
| 13 | 7 | 57 | 120 | Xishan |
| 14 | 4 | 129 | 216 | Putang |
| 15 | 5 | 153 | 180 | Jieshangzu |
| 16 | 3 | 183 | 170 | Jieshangzu |
| 17 | 6 | 110 | 100 | Jieshangzu |
| 18 | 4 | 75 | 80 | Longquanzu |
| 19 | 4 | 173 | 70 | Changguzu |
| 20 | 5 | 200 | 80 | Bamaotian |
| 21 | 5 | 60 | 120 | Bamaotian |
| 22 | 12 | 280 | 320 | Bamaotian |
| 23 | 3 | 82 | 120 | Jianshan |
| 24 | 9 | 80 | 240 | Jianshan |
| 25 | 6 | 45 | 120 | Nongke |
| 26 | 6 | 223 | 110 | Huinson |
| 27 | 5 | 85 | 150 | Fujiazu |
| 28 | 5 | 120 | 100 | Fujiazu |
| 29 | 5 | 134 | 100 | Fujiazu |
| 30 | 6 | 134 | 80 | Fujiazu |
| 31 | 2 | 67 | 150 | Mapozu |
| 32 | 5 | 84 | 100 | Mapozu |
| 33 | 5 | 140 | 170 | Majiazu |
| 34 | 5 | 67 | 150 | Jinjiazu |
| 35 | 2 | 134 | 150 | Maojiazu |
| 36 | 4 | 100 | 130 | Huangguzu |
| 37 | 5 | 335 | 135 | Huangguzu |
| 38 | 6 | 100 | 400 | Huangguzu |

Table 2: Survey data for household vegetable fields in a rural residential area in the Jinjing catchment.

Statistical analysis

All statistical data analyses were performed using R software (<http://www.r-project.org>) with the stats and ade4 packages. For example, descriptive statistical analysis was used to determine the minimum, maximum and mean of the original data. Pearson's correlation analysis was used to evaluate the relationships between daily N₂O fluxes and environmental factors, such as temperatures, precipitation, SWC, and soil NH₄⁺-N and NO₃⁻-N contents. Principal component analysis (PCA) was used to identify the most principal components and explain the internal structure of the environmental variables, and a stepwise regression model and path analysis were used to isolate the most important influencing factors and analyze the interrelationships among those factors, respectively. Origin 8.0 (Origin Lab Ltd., Guangzhou, China) was used to produce some of the graphs. All of the variables were tested for normality; if necessary, natural logarithmic or logit transformation was used for data transformation. In this study, the logit transform of N₂O and the log transforms of NH₄⁺-N and NO₃⁻-N

are denoted as N₂O_t, NH₄⁺-N_t and NO₃⁻-N_t, respectively. Additionally, the Bayesian Markov Chain Monte Carlo (Bayesian MCMC) analysis tool (OpenBUGS, Cambridge Institute of Public Health, UK) was used to calculate the regression equations between the household vegetable field area, the number of family members and the housing area on the basis of our survey data and to estimate the total area of household vegetable fields in the rural residential area in our research catchment.

Results

Precipitation and air temperature

T_{min}, T_{max} and precipitation are shown in Figure 2. The extreme cold days occurred in January in this area, with minimum daily temperatures occurring on 13 January (-4.0°C) and 12 January (-3.7°C) in 2010 and 2011, respectively. The extreme hot days occurred in late July and early August, with T_{max} occurring on 5 August (38.2°C) and 29 July (39.6°C) in 2010 and 2011, respectively. T_{air} ranged from -1.1 to 31.9°C and from -0.5 to 31°C in 2010 and 2011, respectively, with similar fluctuations in T_{min} and T_{max} (data not shown here). Precipitation during the wet season (March to September) accounted for 79.9% and 87.8% of all precipitation in 2010 and 2011 and the greatest monthly precipitation occurred in May in 2010 (296.4 mm) and in June in 2011 (332.6 mm). The annual rainfall was 1,588 mm in 2010 and 1,105 in 2011; the former was greater but the latter was lower than the average annual rainfall (1,330 mm).

Soil temperature and soil water content

As shown in Figure 3a, the T_{soil} at a depth of 5 cm showed same seasonal dynamics as T_{max} and T_{min}; and the lowest and highest T_{soil} occurred in January and August, respectively, in both 2010 and 2011. The SWC at a depth of 0-20 cm showed fluctuation during the two years, from 0.21 ± 0.02 cm³ cm⁻³ to 0.44 ± 0.04 cm³ cm⁻³, with no evident seasonal pattern (Figure 3b). No significant difference in the average SWC was detected between 2010 (0.33 ± 0.03 cm³ cm⁻³) and 2011 (0.32 ± 0.03 cm³ cm⁻³). In the studied household vegetable field, the dry and hot period occurred from late July to early August.

Soil mineral nitrogen contents

The weekly-measured time courses of soil NH₄⁺-N and NO₃⁻-N contents are shown in Figures 3c and 3d. During 2010 and 2011, the soil NH₄⁺-N content ranged from 0.6 to 27.0 mg N kg⁻¹ (averaged at 7.6 mg N kg⁻¹) and from 1.2 to 40.3 mg N kg⁻¹ (averaged at 9.8 mg N kg⁻¹), respectively; while the soil NO₃⁻-N content ranged from 0.1 to 30.2 mg N kg⁻¹ (averaged at 16.2 mg N kg⁻¹) and from 3.0 to 59.1 mg N kg⁻¹ (averaged at 17.0 mg N kg⁻¹), respectively. There was no significant difference in the soil mineral N contents between 2010 and 2011 (*p*>0.05). During the entire observation period, the NH₄⁺-N content, which was lower than the soil NO₃⁻-N content, exhibited troughs from September to November. The soil NO₃⁻-N content exhibited several peaks, but no peaks occurred during the cold winter.

Daily N₂O fluxes and total annual N₂O emissions

Daily N₂O fluxes from the examined household vegetable field in the rural residential area are shown in Figure 3e. In 2010 and 2011, the daily N₂O fluxes ranged from 14.4 to 566.7 μg N m⁻² h⁻¹ (averaged at 175.1 μg N m⁻² h⁻¹) and from 11.5 to 661.1 μg N m⁻² h⁻¹ (averaged at 131.4 μg N m⁻² h⁻¹), respectively. Daily N₂O emissions exhibited evident seasonal dynamics, with emission peaks occurring from March to September and emission troughs occurring from October to the following February. The greatest daily N₂O fluxes occurred on 23 April

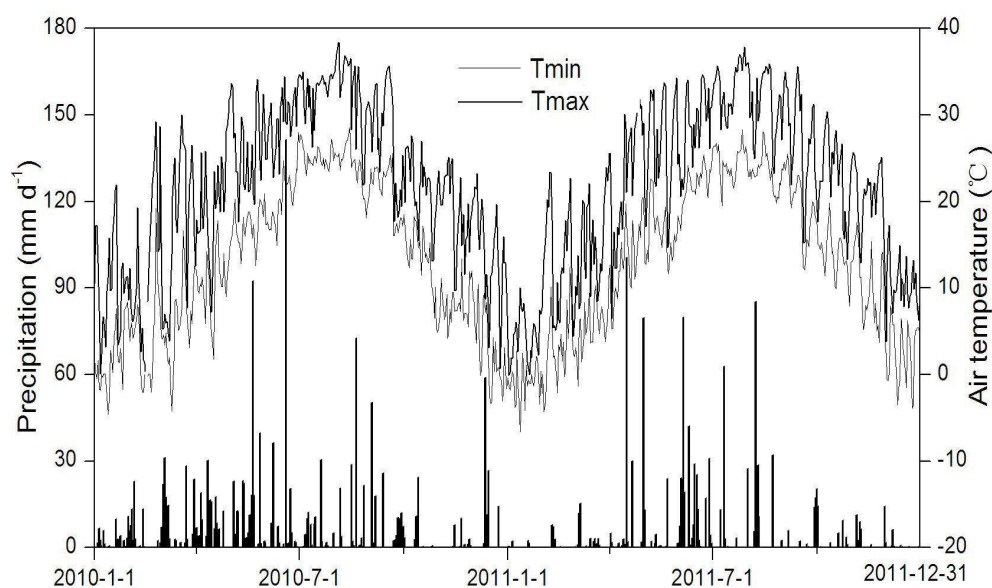


Figure 2: Daily air temperatures and rainfalls.

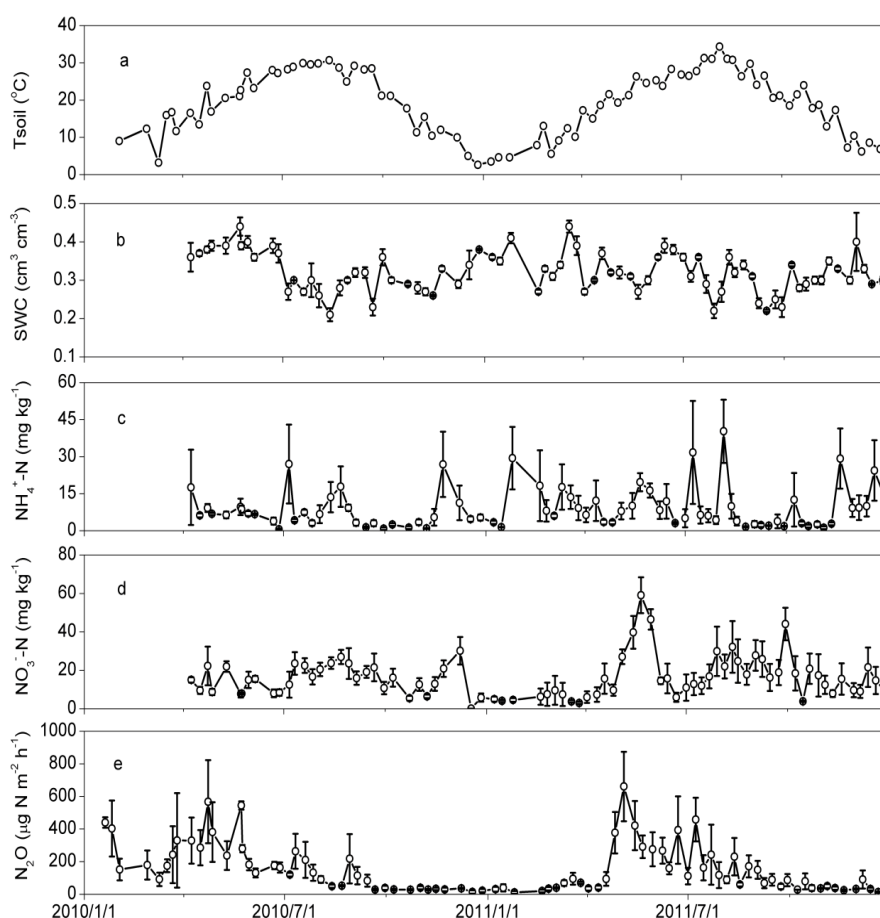


Figure 3: Dynamics of daily N_2O fluxes and soil variables. T_{soil} (a), the soil temperature at a depth of 5 cm; SWC (b), NH_4^+-N (c) and $NO_3^- -N$ (d) indicate the soil volumetric water content, soil ammonium content and nitrate contents of the topsoil at 0–20 cm depth, respectively; N_2O (e) indicates N_2O emissions from a household vegetable field in a rural residential area in the red-soil hilly subtropics of China from 2010 to 2011. The vertical bars indicate the standard errors ($n=3$).

(566.7 $\mu\text{g N m}^{-2} \text{h}^{-1}$) and on 4 May (661.1 $\mu\text{g N m}^{-2} \text{h}^{-1}$) in 2010 and 2011, respectively. The accumulated N_2O emissions in the wet season were significantly greater ($p < 0.05$) than those in the dry season; and former accounted for 77.0% and 90% (averaged at 83.5%) of the total annual N_2O emissions in 2010 and 2011, respectively. The annual N_2O emissions were estimated to be 12.9 ± 2.4 and $11.2 \pm 1.5 \text{ kg N ha}^{-1} \text{ year}^{-1}$ (averaged at $12.1 \pm 0.9 \text{ kg N ha}^{-1} \text{ year}^{-1}$) in 2010 and 2011, respectively; and no significant difference in annual N_2O emissions was detected between the two years ($p > 0.05$).

Relationships between N_2O fluxes and environmental factors

T_{\min} , T_{\max} , T_{air} , T_{soil} , the cumulative precipitation in the previous three days (Rain3t), SWC and $\text{NO}_3^- \text{-Nt}$ were significantly correlated with N_2Ot (Figure 4); and T_{\min} , T_{\max} , T_{air} , T_{soil} and Rain3t displayed very significant, positive correlations with N_2Ot ($r = 0.43, 0.49, 0.47, 0.52$ and 0.41 , respectively; $p < 0.001$). The $\text{NH}_4^+ \text{-Nt}$ showed no significant correlation with N_2Ot in this study ($r = 0.04, p > 0.05$). Moreover, specific correlations were observed between environmental variables. For example, a significant, positive correlation was found between T_{air} and T_{soil} , between $\text{NO}_3^- \text{-Nt}$ and both T_{air} and T_{soil} as well as between SWC and Rain3t, $\text{NO}_3^- \text{-Nt}$, and T_{\max} . The PCA identified seven principal components related to N_2O emissions in the household vegetable field examined in this study (Table 3). The first three principal components accounted for 77.94% of the total variations in environmental variables (temperatures, rainfall, SWC and soil mineral N contents). PC1 was dominated by temperature (T_{\min} , T_{\max} , T_{air} and T_{soil}) and $\text{NO}_3^- \text{-Nt}$; PC2 included precipitation and Rain3t; and PC3 was dominated by SWC. Specifically, the major pattern of N_2O emissions from this field was primarily associated with temperature and soil $\text{NO}_3^- \text{-N}$ content (PC1), followed by rainfall and soil moisture (PC2 and PC3). The result of the stepwise regression analysis confirmed the importance of the correlated factors of T_{soil} and T_{\min} , as well as soil moisture and Rain3t. The generated model was as follows:

$$\text{N}_2\text{Ot} = -10.2 + 0.230T_{\text{soil}} + 9.84\text{SWC} + 2.18\text{Rain3t} - 0.128T_{\min} \quad (1)$$

This model, which used the four continuous variables T_{soil} , T_{\min} , SWC and Rain3t, exhibited great performance ($R^2_{\text{adj}} = 0.48$, $\text{RMSE} = 1.14$; $p < 0.001$) and explained 48.1% of the total variation in the logit-transformed daily N_2O fluxes from the examined household vegetable field. Path analysis also indicated that T_{soil} was the most significant positive factor for N_2O emissions (Table 4); T_{\min} influenced N_2O negatively and indirectly by controlling T_{soil} ; and higher T_{soil} confounded with higher soil moisture enhanced N_2O emissions.

Survey and estimation of the total household vegetable field area

A model was constructed according to the Bayesian MCMC method using information from 38 surveyed rural families. The obtained model was as follows:

$$\frac{Y}{10} = A_0 + A_1 \times FM + A_2 \times \frac{HA}{75} + b[i] \quad (2)$$

where Y refers to the vegetable field area per family, FM refers to the number of family members per family, HA refers to the housing area per family, and $b[i]$ denotes the statistical error, which displayed a normal distribution. The constants of 10 and 75 were used to normalize the original data for Y and HA to facilitate the generation of the model. The model showed great performance, as $A_0 = 3.28$, $A_1 = 0.778$ and $A_2 = 2.11$ ($\text{sd} = 2.61, 0.504$ and 0.119 , respectively; $\text{MC-error} = 0.101, 0.019$ and 0.001 , respectively). In the study catchment, the total housing area was 342.11 ha and the total population was 0.45 million; therefore, the

estimated total household vegetable field area was $130.8 \pm 0.56 \text{ ha}$.

Estimation of the annual total N_2O emissions from the household vegetable fields in the study catchment

Based on the estimated total household vegetable field area of $130.8 \pm 0.56 \text{ ha}$ in the study catchment, the annual total N_2O emissions from these fields were $1.69 \pm 0.01 \text{ ton N year}^{-1}$ and $1.46 \pm 0.01 \text{ ton N year}^{-1}$ in 2010 and 2011, respectively (Table 5).

Discussion

Seasonal dynamics of N_2O emissions

The continuous observations suggested that most of the N_2O emissions were emitted during the wet season (83.5% of annual N_2O emissions), mainly due to the increases in temperature and soil moisture resulting from heavy rain. Similar results have been reported previously [24–26]. In the wet and warm season, greater N_2O emissions occurred with more tillage activities, such as transplanting, fertilization, and irrigation [9,27,28]. In particular, liquid human excreta application was performed from late March to August (growing seasons) and these applications induced several N_2O emission peaks during the wet and warm season, especially from April to July. When the weather became colder, new rotations stopped but fertilization activities continued. The lower T_{\min} and T_{soil} restricted microbial activities and depressed N_2O production in the soil [29], resulting in lower N_2O emission peaks in the household vegetable field. The same phenomenon was observed in other studies [10]. These results implied that the control on N_2O emissions during the wet and warm season is very important for mitigating greenhouse gas emissions from household vegetable fields in rural residential areas.

Impacts of environmental factors on N_2O emissions

The results of the stepwise regression model (Eq. 1) and the path analysis (Table 4) indicated that soil temperature and soil moisture were the most important environmental factors controlling N_2O emissions in the examined vegetable field. In this study, significantly greater N_2O emissions were observed in warm and rainy months than in cold months, consistent with previous studies [11,30]. The rainfall directly influenced the soil moisture and the soil moisture fluctuation exerted an important effect on the quantity of microbes in soil, the substrate utilization pattern and the microbial community composition [31,32]. In addition, frequent liquid human excreta application has been reported to lead to frequent wetting-drying cycles in the topsoil, increasing the net soil N mineralization, microbial activity and microbial biomass in greenhouse vegetable soils [33]. In this study, the large N_2O emissions from the household vegetable field can be attributed to the specific N application method because only liquid human excreta were applied freely.

Soil $\text{NH}_4^+ \text{-N}$ and $\text{NO}_3^- \text{-N}$ are necessary substrates for nitrification and denitrification processes [34]. In this study, $\text{NO}_3^- \text{-N}$ and temperature were included in the first principal component and the results of stepwise regression analysis indicated that temperature and soil moisture were the most important controlling factors for N_2O emissions from this soil. In addition, the high-frequency application of liquid excreta resulted in the application of more active N, and the soil $\text{NO}_3^- \text{-N}$ content was always higher than 5 mg kg^{-1} (Figure 3d). These factors indicate that the soil mineral content should not be a limiting factor in N_2O emissions. Moreover, the wetting-drying cycle resulting from frequent liquid human excreta application stimulated

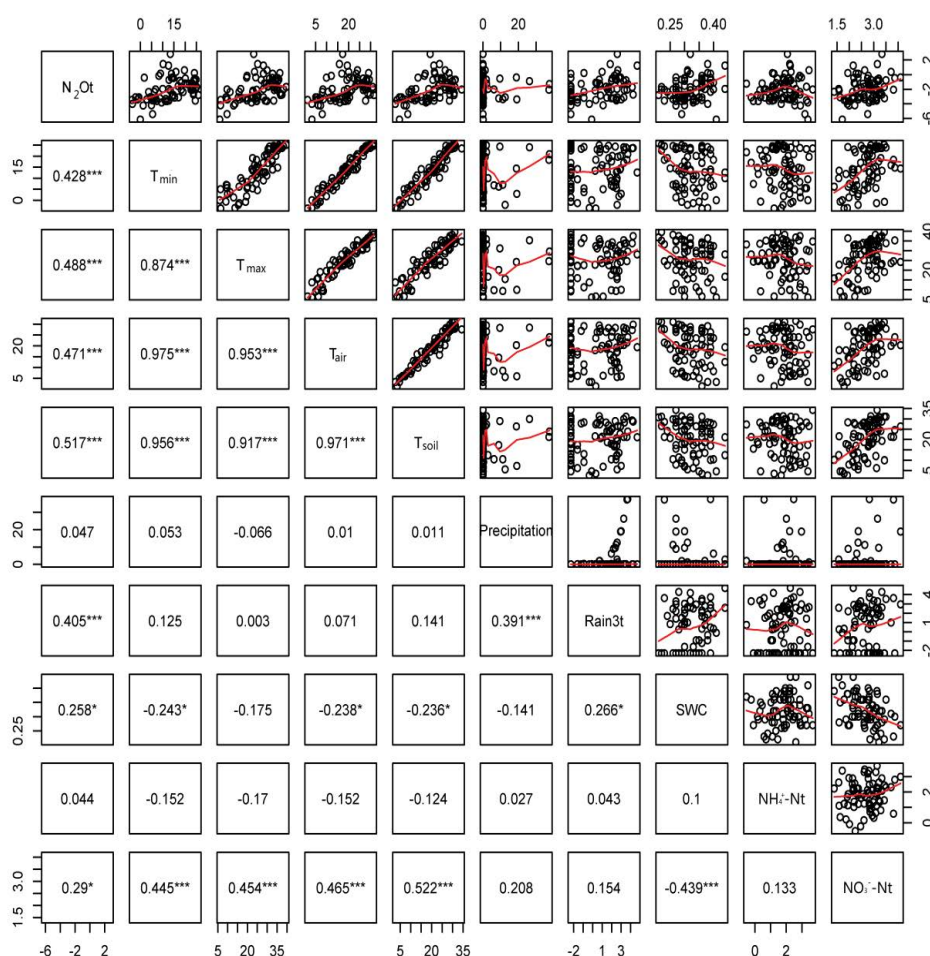


Figure 4: Coefficient matrix of Pearson's correlation of N_2O emissions and environmental factors. All variables in the correlation matrix were normally distributed. N_2O represents the N_2O flux ($\mu g N m^{-2} h^{-1}$); T_{min} , daily minimum temperatures ($^{\circ}C$); T_{max} , daily maximum temperatures ($^{\circ}C$); T_{air} , daily mean temperatures ($^{\circ}C$); T_{soil} , soil temperature at 0–20 cm depth ($^{\circ}C$); Precipitation, the daily precipitation ($mm d^{-1}$); Rain3t, the cumulative precipitation of the previous three days (mm); SWC, the soil volumetric water content (cm^3); NH_4^+-N , the soil ammonium content ($mg N kg^{-1}$); $NO_3^- - N$, the soil nitrate content ($mg N kg^{-1}$). *, ** and *** indicate statistical significance at probability levels of 0.05, 0.01 and 0.001. The subscript of t represents the logit or log transformation.

household vegetable field in the rural residential area examined in this study. Although a reduced denitrification rate, N_2O/N_2 ratio and N_2O emissions were reported for an intensive vegetable field with pig manure applications [28,36] other studies have observed increased N_2O emissions with manure applications compared with only chemical fertilizer application and significantly greater N_2O emissions following human excreta application [11]. In this study, human excreta, which contains large amounts of soluble N, DOC and water, were applied as a basic fertilizer to the household vegetable field and promoted microbial activities and N_2O emissions. Therefore, performing observations at a high frequency (2 to 3 times a week) or even utilize a continuous measurement system is necessary to produce more accurate results.

N_2O re-emissions of the input nitrogen into the catchment

The IPCC [1] indicated that approximately 0.2 Tg (range 0.1–0.3 Tg) of N_2O-N were derived from human excreta globally in 2006. This estimate was insufficient to accurately estimate the total N_2O emissions originating from human excreta at the catchment scale because of differences in geographic position, economic development levels,

lifestyle, diet, etc. In this study, we assumed that 6.19 kg N per capita [22] is returned to the environment in the Jinjing catchment, ignoring differences in sex, age, diet, etc. Accordingly, the total N input resulting from human excreta was 278.6 ton $N year^{-1}$ in the studied basin. N_2O emissions from household vegetable soils to which only human excreta were applied were estimated to be 1.58 ± 0.16 ton $N year^{-1}$. Thus, N_2O emissions from household vegetable fields in rural areas are likely categorized as direct N_2O emissions, as described in IPCC [40]. However, such source of N_2O should be categorized as N_2O re-emissions when estimating the regional N_2O inventory in a catchment to avoid double-counting the N input and underestimating N_2O-N export. N is consumed daily by humans through food, including grain, meat, eggs and vegetables and leaves the soil ecosystems. A portion of that N is returned to the environment as human excreta under different conditions, such as to the soil as organic fertilizer or to rivers and ponds via sewage systems as sewage waste. During the N cycling in this process, N_2O is produced as an intermediate product and emitted into the atmosphere. Thus, defining such N_2O emissions as N_2O re-emissions at a regional scale is more appropriate. Unfortunately, N_2O re-emissions

| Factor | PC1 | PC2 | PC3 | |
|----------------------------------|--------|--------|--------|------------------------|
| T _{min} | 0.958 | -0.023 | 0.120 | |
| T _{max} | 0.938 | -0.154 | 0.148 | |
| T _{air} | 0.980 | -0.073 | 0.119 | |
| T _{soil} | 0.977 | -0.015 | 0.106 | |
| Precipitation | -0.019 | 0.823 | -0.128 | |
| Rain3t | 0.152 | 0.797 | 0.415 | |
| SWC | -0.343 | 0.007 | 0.856 | |
| NH ₄ ⁺ -Nt | -0.176 | 0.307 | -0.159 | |
| NO ₃ ⁻ -Nt | 0.616 | 0.330 | -0.445 | |
| | 4.264 | 1.545 | 1.206 | Standard deviation |
| | 47.381 | 17.164 | 13.398 | Proportion of variance |
| | 47.381 | 64.544 | 77.94 | Cumulative proportion |

Note: T_{min}, daily minimum temperatures (°C); T_{max}, daily maximum temperatures (°C); T_{air}, daily mean temperatures (°C); T_{soil}, soil temperature at 0–20 cm depth (°C); Precipitation, the daily precipitation (mm d⁻¹); Rain3, the cumulative precipitation in the previous three days (mm); SWC, the soil volumetric water content (cm³). The subscript of t represents the logit or log transformation.

Table 3: Principal component analysis of environmental factors influencing N₂O emissions.

| Factor | Direct path coefficient | Indirect path coefficient | | | | Total |
|-------------------|-------------------------|---------------------------|--------|--------|------------------|--------|
| | | T _{soil} | SWC | Rain3t | T _{min} | |
| T _{soil} | 1.215 | - | -0.078 | 0.038 | -0.657 | -0.698 |
| SWC | 0.328 | -0.289 | - | 0.049 | 0.167 | -0.072 |
| Rain3t | 0.229 | 0.200 | 0.070 | - | -0.102 | 0.169 |
| T _{min} | -0.687 | 1.163 | -0.080 | 0.034 | - | 1.117 |

Note: T_{min}, daily minimum temperatures (°C); T_{soil}, soil temperature at 0–20 cm depth (°C); Rain3, the cumulative precipitation in the previous three days (mm); SWC, the soil volumetric water content (cm³). The subscript of t represents the log transformation.

Table 4: Path coefficients of environmental factors influencing N₂O emissions.

| Year | N ₂ O emissions (kg N ha ⁻¹ year ⁻¹) | Basin area (km ²) | Area of household vegetable fields (ha) | Total N ₂ O emissions (ton N year ⁻¹) |
|------|--|-------------------------------|---|--|
| 2010 | 12.9 ± 2.4 | 135 | 130.8 ± 0.56 | 1.69 ± 0.01 |
| 2011 | 11.2 ± 1.5 | 135 | 130.8 ± 0.56 | 1.46 ± 0.01 |

Table 5: Annual N₂O emissions from the household vegetable fields in the Jinjing catchment.

soil N mineralization from organic N fertilizer [32,33] and subsequent vegetable cultivation improved soil NO₃⁻-N accumulation, resulting in greater N₂O emissions under conditions of suitable soil moisture [35]. In this study, the main reason for the similar soil NO₃⁻-N content between 2010 and 2011 was the large amount of N lost by leaching. N leaching is very significant in red soils in subtropical China because of the high N applications and high rainfalls, particularly during the wet and warm season. The high frequency of cultivating activities further aggravated N leaching. Thus, there was no significant difference in soil NO₃⁻-N content between 2010 and 2011. However, the soil NO₃⁻-N content was relatively high (greater than 5 mg N kg⁻¹) in this study. Pearson's correlation analysis also revealed a significant, positive correlation between N₂O_t and NO₃⁻-N_t ($r=0.29$, $p<0.05$), implying that the denitrification process is the dominant contributor to N₂O emissions in the examined soil. Additionally, several previous studies have reported that organic excreta application to the soil may provide more dissolved organic carbon (DOC), which enhances the activities of denitrifiers and transforms more available N into N₂O, thereby increasing soil N₂O emissions [11,36]. In household vegetable fields, the high frequent application of liquid excreta, a type of organic fertilizer, also supplied abundant DOC for microbial activities that promote soil N₂O emissions.

Comparison with other land use types

In this study, the average annual N₂O emissions were 12.1 ± 0.9 kg N ha⁻¹ year⁻¹ for 2010–2011 from the examined household vegetable field in the rural residential area, which had a total N application of 619 kg N ha⁻¹ year⁻¹. These emissions are similar to 12.0 kg N ha⁻¹ from open-

air intensive vegetable fields with higher total N application (1,636 kg N ha⁻¹) during a longer period (14 months) reported by Xiong et al. [18] and lower than 16.5 kg N ha⁻¹ under conventional chemical fertilizer N treatment consisting of 1,710 kg N application observed by He et al. [9]. However, the results of the present study are comparable to those for other open-air intensive vegetable and greenhouse vegetable cultivation systems [16]. Consistent with previous reports, the annual N₂O emissions from the examined vegetable field soil were much greater than those from native grassland, forest and other cropland soils [25,26,37]. In the studied catchment, the annual N₂O emissions from the examined household vegetable field in the rural residential area were eightfold greater than those from a Masson pine forest soil (1.50 kg N ha⁻¹ year⁻¹) [38] and two-thirds of those from a tea field soil (17.2 kg N ha⁻¹ year⁻¹) in the same study area [23]. Although the weekly measurements may have slightly overestimated the annual N₂O emissions because of the large temporal and spatial variations in N₂O fluxes [27], the results of our study suggest that household vegetable fields in rural residential areas of subtropical China may be a hot-spot of greenhouse gas emissions, even though this land constitutes a relatively small portion of croplands. For example, in 2011, the total N₂O emissions from household vegetable fields (0.48% of the Jinjing catchment) were 0.72 ton N year⁻¹, whereas, the total N₂O emitted from paddy fields (24.8% of the Jinjing catchment) was estimated to be 0.52 ton N year⁻¹ based on Liu et al. [39].

The greater N₂O emissions might be attributable to the application of liquid organic excreta, which was the only fertilizer source in the

from the input N originating from human excreta were measurable only in household vegetable fields at the Jinjing catchment scale and did not represent the entire N₂O emissions related to excreta because there are several alternative pathways of the release of human excreta, such as release into rivers, ponds or other crop soils. Furthermore, management decisions regarding excreta disposal and storage affect N₂O emissions and flush toilets and septic tanks may utilize distinct N cycling pathways that induce N₂O emissions. Thus, the emission factor of N₂O re-emissions (0.57%) estimated in this study implies that the N₂O re-emissions from household vegetable fields originating from N input via human excreta were unnegligible.

Conclusion

Based on two years of *in situ* continuous measurements in a household vegetable field of a rural residential area in subtropical central China, the annual N₂O emissions were estimated to be 12.9 ± 2.4 kg N ha⁻¹ year⁻¹ and 11.2 ± 1.5 kg N ha⁻¹ year⁻¹ in 2010 and 2011, respectively. No significant difference in annual N₂O flux was detected between the two years. The total N₂O emissions from household vegetable fields in the rural residential area of the studied Jinjing catchment were 1.69 ± 0.01 ton N year⁻¹ and 1.46 ± 0.01 ton N year⁻¹ in 2010 and 2011, respectively. The seasonal pattern of N₂O emissions was primarily influenced by temperature and SWC, especially the latter, as a high soil mineral N content resulted from frequent application of liquid human excreta. These results suggest that denitrification was responsible for the high N₂O emissions from this field. Consistent with the findings in other studies, the household vegetable field in the rural residential area was a source of atmospheric N₂O and was even a potential hot-spot of N₂O emissions in the subtropical region of China. Exclusive, intensive application of liquid human excreta directly to the soil may explain the high N₂O emissions from these fields. These findings suggest that sustainable management practices are needed to reduce the environmental risk of human excreta, particularly induced N₂O emissions. At the catchment scale, a relevant level of N₂O re-emissions was released due to human excreta application on household vegetable fields, and additional N₂O emission sources, such as water bodies (e.g. ponds, lakes and streams) and septic tanks themselves, must be explored.

Authors' Contribution

XF conducted all laboratory experiments and drafted the manuscript with YL. YL finalized the manuscript. All authors designed the field experiment, participated in the data analysis and reviewed and approved the final manuscript.

Competing Interests

All authors declare that they have no competing interests.

Acknowledgement

This research was financially supported by the National Basic Research Program of China (2012CB417105).

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Citation: Fu X, Li Y, Wang Y, Shen J, Xiao R, et al. (2016) Nitrous Oxide Emissions from Household Vegetable Fields in the Rural Residential Areas of Hilly Subtropical Central China. *Environ Pollut Climate Change* 1: 103. doi: [10.4172/2573-458X.1000103](https://doi.org/10.4172/2573-458X.1000103)