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Effects of N Fertilizer Application on Soil N_2O Emissions and CH_4 Uptake: A Two-Year Study in an Apple Orchard in Eastern China

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Abstract: Land use changes from cropland to orchards in Eastern China have raised serious concerns about the regional nitrogen (N) cycle and greenhouse gas balance. We measured soil nitrous oxide (N_2O) emissions and methane (CH_4) uptake using manual static chambers in an apple orchard. The primary aims were to assess the effect of N fertilizer application on gas fluxes and quantify the site-specific N_2O emission factor (EF_d). Field experiments were arranged in a randomized block design with three N input rates (0, 800 and 2600/2000 kg N ha^{-1} $year^{-1}$). We found that orchard soils were a negligible CH_4 sink (−1.1 to −0.4 kg C ha^{-1} $year^{-1}$). Annual N_2O emissions responded positively to N input rates, ranging from 34.1 to 60.3 kg N ha^{-1} $year^{-1}$. EF_d ranged from 1.00% to 1.65% with a mean of 1.34%. The extremely large background emissions of N_2O (34.1–34.3 kg N ha^{-1} $year^{-1}$) most likely originated from nitrate accumulation in the soil profile because of historical overuse of N fertilizer. We conclude that (1) site-specific EF_d is suitable for assessing regional direct N_2O emissions from upland orchards; and (2) conventional fertilization regimes must be avoided, and reduced N input rates are recommended in the study region.

Keywords: greenhouse gas; direct emission factor; background emissions; fertilization; production; upland orchard

1. Introduction

Nitrous oxide (N_2O) and methane (CH_4) are potent greenhouse gases. Together, they are responsible for 30% of global radiative forcing [1]. Atmospheric concentrations of N_2O and CH_4 have increased by 20% and 150%, respectively, since 1750, most likely due to human activities related to agriculture [1]. Reducing N_2O emissions from and enhancing CH_4 uptake by upland agricultural ecosystems could help mitigate anthropogenic contributions to global warming [2].

Agricultural soils are responsible for 60–80% of anthropogenic N_2O sources [3,4]. Soil N_2O is primarily produced through microbial processes such as nitrification, denitrification, and nitrifier denitrification, which may occur simultaneously in soil aggregates if inorganic nitrogen (NH_4^+ and NO_3^-) substrates are available and soils are aerated [5,6]. Denitrification becomes the major source of

N_2O when oxygen concentrations in soil air are limited, primarily when a soil's water-filled pore space (WFPS) is greater than 60% [7,8]. In field measurements, large N_2O emissions usually occur when nitrogen (N) fertilizer applications are accompanied by rainfall or irrigation events [9–14]. Annual N_2O emissions have been shown to increase with N input rates; this fact led to the development of the direct emission factor (EF_d) concept [15–17]. EF_d is the fraction of N fertilizer lost as N_2O for a given year or season and has been widely used to calculate global and regional N_2O emissions [3,4,18]. To compile national N_2O emissions inventories, IPCC Guidelines [19] provide a default EF_d of 1% and a large uncertainty range (from 0.3% to 3%) for cultivated mineral soils. Such uncertainty is primarily due to the complex relationships between N_2O emissions and soil properties, climate, and artificial practices [11,12,20,21]. Although the N_2O flux data recorded has increased in recent years, there is still a great deal of uncertainty in the assessment of EF_d, and therefore N_2O, emissions [15,18,20]. The quantification of site-specific EF_d is critical to the accurate assessment of N_2O emissions in a given region [18,21,22].

Agricultural activities, notably fermentative digestion by ruminant livestock, manure management, and rice grown under flooded conditions, account for approximately 60% of anthropogenic CH_4 emissions [1]. Upland agricultural soils also remove CH_4 from the atmosphere through oxidation by aerobic methanotrophic bacteria, but this effect is small. Large soil water and NH_4^+ quantities significantly inhibit CH_4 uptake [23,24].

In Eastern and Northwestern China, large amounts of cropland have been converted to orchards and vegetable fields for economic benefits [25]. Together, these two regions account for 87.8% and 88.7% of the national apple acreage and yield, respectively [26]. This land use change raises serious concerns about the regional N cycle and greenhouse gas balance [27,28]. In Eastern China, the mean N application rate was 603 kg N ha^{-1} $year^{-1}$ for cropland in 2006, while the rate was an average 842 and 3239 kg N ha^{-1} $year^{-1}$ for orchards and greenhouse vegetables, respectively [27]. Previous research has revealed that the overuse of N fertilizer leads to excessive N substrates in the soil profile beneath cereals and is associated with NO_3^- leaching and N_2O emissions [9,29,30]. However, the impact of intensive N management on N_2O and CH_4 fluxes in upland orchards is not well understood because quantitative information on this topic is scarce. Reported N_2O emissions from orchard soils range from 0.5 to 8.6 kg N ha^{-1} $year^{-1}$ and vary in response to climate, land use, and management practices [14,31–35]. These observations are similar to most cropland-related N_2O emissions reviewed in Stehfest and Bouwman [20]. These results are reasonable because the N input rates analyzed in these studies were largely within 200–300 kg N ha^{-1} $year^{-1}$, which is similar to input rates in most croplands. However, it is unclear whether these previous observations can be extrapolated more generally to Eastern China due to the extremely large N input rates.

With respect to local soil management, we hypothesized that annual N_2O losses and EF_d were significant in an upland orchard in Eastern China. We measured soil N_2O emissions and CH_4 uptake regularly in an irrigated apple orchard between 2010 and 2011. The primary aims of this study were to assess the effect of N input rates on gas fluxes and quantify site-specific EF_d.

2. Experiments

2.1. Site Description

The experimental sites were located in an orchard (37°49′ N, 120°45′ E) dominated by Fuji apple trees in the suburb of Penglai County, Shandong province, one of the major apple-producing regions in Eastern China. This region is characterized by a warm, temperate, continental monsoon climate, with a mean long-term (1961–1990) annual precipitation value of 664 mm, an annual mean air temperature of 11.9 °C, and an annual frost-free period of 206 days. The orchard is approximately 2 ha square and was converted from a winter wheat field in 2002. Apples were first harvested from the orchard in October 2005. The tree density was 620 tree ha^{-1}, and the size of the canopy of an adult apple tree was approximately 16 m^2. Topsoil properties (0–20 cm layer) at the beginning of the study (October 2009) are listed in Table 1.

Table 1. Selected soil properties (0–20 cm layer).

Soil Properties	Mean
pH (H_2O)	6.7
Bulk density (g cm^{-3})	1.39
Soil organic carbon (g C kg^{-1})	10.6
Total nitrogen (g N kg^{-1})	1.2
Total phosphorus (g P kg^{-1})	1.9
Total potassium (g K kg^{-1})	15.8
Available phosphorus (mg P kg^{-1})	50.7
Available potassium (mg K kg^{-1})	155.2
Clay (<2 μm, %)	7.0
Silt (2–200 μm, %)	39.6
Sand (>200 μm, %)	53.4

2.2. Experimental Design

Field measurements were conducted from January 2010 to December 2011. Three N fertilizer rate treatments for each year (0, 800, and 2600 kg N ha^{-1} $year^{-1}$ in 2010 and 0, 800, and 2000 kg N ha^{-1} $year^{-1}$ in 2011, hereafter referred to as CK, LN, and HN, respectively) were arranged in a randomized block design. Each treatment was replicated three times. HN represents the conventional treatment or sites where the soils have been under similar management since 2002. LN represents sites where the recommended management practice was instituted. In both the LN and HN treatments, urea was applied in separate applications of 50%, 15%, and 35%, the first in early April (basal application), the second in late June (first topdressing), and the last in late August (second topdressing), respectively. The basal fertilizers were buried in ditches (length × width × depth = 1.0 m × 0.3 m × 0.2 m) that were evenly arranged around each apple tree like a cross. The positions of the fertilizer ditches were changed every year. The dressing fertilizers were applied at the surface under the tree canopies one day after a rainfall or irrigation event. The proportion of the fertilizer ditches and bare soil areas at the field scale was 15% and 85%, respectively. All three treatments received a single application of phosphorus (400 kg P_2O_5 ha^{-1} $year^{-1}$) and two separate applications of potassium (800 kg K_2O ha^{-1} $year^{-1}$). The soils were flood irrigated with underground water three to five times each year, depending on the precipitation pattern. The water input rate for each irrigation event was similar to a rainfall event of 40 mm. More information about the fertilization and irrigation regimes can be found in Table 2. Pesticides and fungicides were applied as a foliar spray two or three times each year to control pests and diseases.

Table 2. Management practices in the apple orchard between 2010 and 2011.

Year	Fertilization Date	Method	Fertilizer	Date of Irrigation
2010	1 April	Buried in ditches	50% N 100% P 50% K	11 May; 2 August; 1 October; 13 November; 7 December
	24 June	Surface application	15% N	
	22 August	Surface application	35% N 50% K	
2011	1 April	Buried in ditches	50% N 100% P 50% K	27 March; 20 April; 27 May
	24 June	Surface application	15% N	
	22 August	Surface application	35% N 50% K	

Treatment CK was not fertilized with N fertilizer in the studied years. Treatment LN was fertilized with urea at 800 kg N ha^{-1} $year^{-1}$ in both years. Treatment HN was fertilized with urea at 2600 and 2000 kg N ha^{-1} $year^{-1}$ in 2010 and 2011, respectively. All treatments were fertilized with phosphorus (400 kg P_2O_5 ha^{-1} $year^{-1}$) and potassium (800 kg K_2O ha^{-1} $year^{-1}$) and flood irrigated.

2.3. Measurement of N_2O and CH_4 Fluxes

Manual static chambers were used to measure gas fluxes [36]. Considering the spatial heterogeneity of the basal fertilizer applications, we sampled gases at both fertilizer ditches (FD) and the bare soil (BS) under selected tree canopies in each replicate. A stainless-steel frame (length × width × depth = 0.5 m × 0.5 m × 0.2 m) was permanently inserted approximately 15 cm deep in the soil at each sampling site. During gas flux measurements, a stainless-steel chamber (length × width × height = 0.5 m × 0.5 m × 0.5 m) was mounted on the frame for 30 min, and five gas samples (20 mL) were taken with plastic syringes at 6 min intervals. The sampling process was always completed between 8:00 and 10:00 (local time) on each sampling date, and the sites were monitored in a random order. Gases were usually sampled once a week and up to four times a week for one week after each fertilization event. Previous research has shown that weekly N_2O flux measurements complemented by additional event-related flux determinations provide accurate annual emission estimates [10,37,38]. N_2O and CH_4 concentrations were analyzed within 12-h of sampling using a gas chromatograph (7890A, Agilent Technologies Inc., Santa Clara, CA, USA) equipped with an electron capture detector and a hydrogen flame ionization detector [39].

Gas flux was calculated from the initial change rate of N_2O and CH_4 concentrations during deployment in the chamber [40]. Only when the linear regression of the gas concentrations and the enclosure time was statistically significant at $p < 0.05$ was the initial change rate was accepted to calculate a valid flux value. Annual N_2O emissions and CH_4 uptake were estimated via the linear interpolation of data points. Fluxes were extrapolated to the field scale using the ratio of FD to BS areas (0.38). EF_d was the fraction of the differences between annual N_2O emissions from the fertilized and unfertilized treatments to the N input rate at the field scale in the analyzed year.

2.4. Ancillary Measurements

Local weather conditions were provided by the China Meteorological Administration. Soil temperatures and volumetric soil water content (VSWC) at 10 cm depth were measured automatically once an hour (SMSC04 soil moisture station, Beijing Unism Technologies Inc., Beijing, China) across a 100 m^2 region. Daily soil temperature and VSWC were calculated as the mean of the time series for the three replicates and presented as the mean condition within the experimental orchard. The VSWC measurements were converted to WFPS with an assumed soil particle density of 2.65 g cm^{-3}.

In 2010, three augers of soil (0–10 cm layer) were randomly collected near each chamber once every two weeks and mixed into one sample until thoroughly combined (approximately 1–2 kg of fresh soil). The fresh soil samples were divided into two sub-samples. One sub-sample (5 g) was extracted with deionized water (soil:water = 1:5). The extracts were immediately filtered through a polyethersulfone membrane (45 μm pore size) and analyzed for dissolved organic carbon (DOC) using an organic carbon analyzer (TOC-5000A, Shimadzu, Kyoto, Japan). The other sub-sample (5 g) was extracted with a KCl solution (2 mol L^{-1}) to determine the NH_4^+ and NO_3^- contents using an automated discrete photometric analyzer (AA2, BRAN & Lubbe, Nordstedt, Germany).

Towards the end of October in each year, we harvested and weighed fresh apples from randomly selected trees in each treatment. The orchard's production was extrapolated to the field scale using the tree density.

2.5. Statistical Analysis

Statistical analyses were performed using SYSTAT for Windows (version 12.0 Systat Software Inc., Chicago, IL, USA). Differences in N_2O and CH_4 emissions and soil parameters among various

treatments were determined statistically using one-way ANOVA analysis. Linear regressions were used to assess the relationships between gas fluxes and selected soil parameters.

3. Results

3.1. Climate, Soil Temperature, and WFPS

The annual mean air temperature was approximately 12.6 °C in both years (Figure 1a). Annual precipitation rates in 2010 and 2011 were 13% higher and 9% lower than the long-term historical average, respectively. More than 85% of the precipitation in the area occurred from May to October in both years. Soil temperature (10 cm depth) ranged from −3.2 °C to 27.3 °C during the experimental period, with a mean of approximately 9.2 °C in both years (Figure 1b). Soil WFPS (10 cm depth) was mostly above 60% during the wet season and averaged 64% over the entire experimental period.

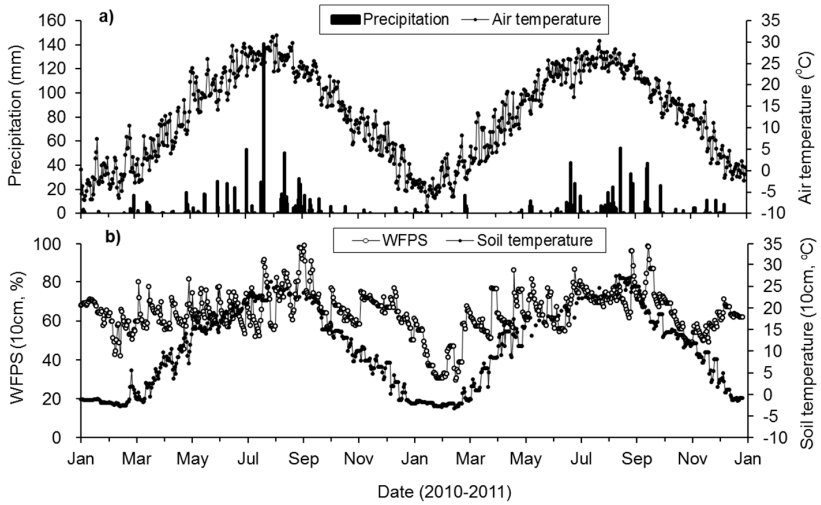

Figure 1. Seasonal dynamics between (**a**) air temperature (°C) and precipitation (mm); and (**b**) soil temperature (10 cm depth, °C) and water-filled pore space (WFPS; 10 cm depth, %) in the studied apple orchard during 2010–2011.

3.2. Soil DOC, NH_4^+ and NO_3^- Content

Soil DOC values were largely within 100–200 mg C kg^{-1} across all treatments and sampling sites (Figure 2) with relatively small temporal variations (CV ranging from 12% to 22%).

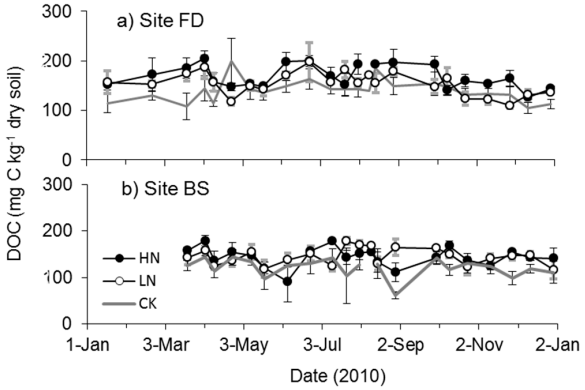

Figure 2. Seasonal dynamics in dissolved organic carbon (DOC) content (mg C kg^{-1} dry soil) across the sampling sites and treatments in 2010. (**a**) Fertilizer ditches; (**b**) Bare soil; Data points indicate the mean of the three replicates; error bars indicate the standard error (SE).

Soil NH_4^+ content was between 6.4–47.3, 9.0–206.0, and 11.2–200.9 mg N kg^{-1} for the CK, LN, and HN treatments, respectively (Figure 3). The NH_4^+ contents from the fertilized treatments were significantly larger than CK treatments ($p < 0.01$). At the BS site, soil NH_4^+ content was between 2.3–8.6, 2.9–47.5, and 3.2–122.7 mg N kg^{-1} for the CK, LN, and HN treatments, respectively (Figure 3). The NH_4^+ contents from HN treatments were significantly larger than those from the LN ($p < 0.05$) and CK treatments ($p < 0.01$).

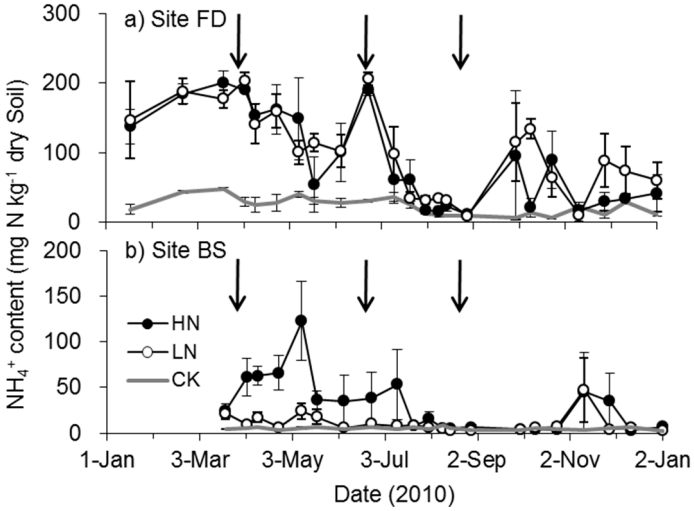

Figure 3. Seasonal dynamics of soil NH_4^+ content (mg N kg^{-1}) across the sampling sites and treatments in 2010. (**a**) Fertilizer ditches; (**b**) Bare soil; Data points indicate the mean of the three replicates; error bars indicate the standard error (SE). Black arrows indicate nitrogen fertilizer applications.

Soil NO_3^- content at the FD site was between 4.3–41.8, 5.6–143.1, and 4.1–147.1 mg N kg^{-1} for the CK, LN, and HN treatments, respectively (Figure 4). At the BS site, soil NO_3^- content was between 6.6–29.7, 9.7–80.0, and 8.3–119.8 mg N kg^{-1} for the CK, LN, and HN treatments, respectively. The NO_3^- contents from the fertilized treatments were significantly larger than CK treatment at both FD and BS sites ($p < 0.05$).

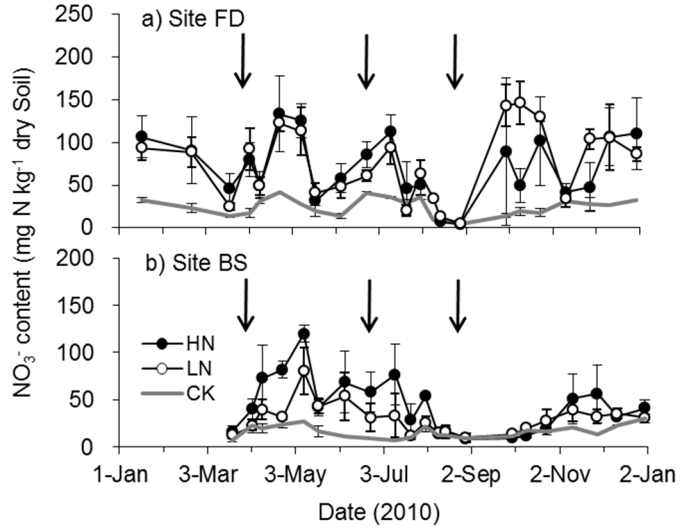

Figure 4. Seasonal dynamics of soil NO_3^- content (mg N kg^{-1}) across the sampling sites and treatments in 2010. (**a**) Fertilizer ditches; (**b**) Bare soil; Data points indicate the mean of the three replicates; error bars indicate the standard error (SE). Black arrows indicate nitrogen fertilizer applications.

3.3. Apple Production

Apple production ranged from 26.2 to 35.6 t ha^{-1} (n = 6, CV = 11%) across the treatments and experimental years (Table 3). Production in the fertilized treatments tended to be larger (ranging from 14% to 19%) than that in the unfertilized treatment areas; however, the differences were not significant ($p > 0.05$) in most cases. Apple production in the LN treatment was 32% higher than the CK treatment in 2011 ($p < 0.01$).

Table 3. Apple production, annual N_2O emissions, EFd, and CH_4 uptake during 2010 and 2011.

Year	Treatment	Production (t ha^{-1})	N_2O (kg N ha^{-1} Year^{-1})			EF_d (%)	CH_4 (kg C ha^{-1} Year^{-1})
			FD	BS	Field		Field
2010	CK	26.2 (2.4)a	39.1 (2.0)a	32.1 (3.5)a	34.1 (3.1)a	-	−0.9 (0.4)a
	LN	31.2 (5.1)a	68.2 (13.7)ab	39.2 (1.9)ab	47.2 (5.2)b	1.65	−1.1 (0.6)a
	HN	30.2 (0.8)a	93.6 (11.3)b	47.6 (2.9)b	60.3 (5.2)c	1.46	−0.4 (0.7)a
2011	CK	27.1 (2.6)a	43.1 (1.1)a	30.9 (3.4)a	34.3 (2.7)a	-	−1.1 (0.6)a
	LN	35.6 (1.8)b	77.1 (12.3)ab	33.7 (3.8)a	45.7 (6.1)b	1.00	−1.1 (0.4)a
	HN	30.9 (3.3)ab	112.7 (14.2)b	38.7 (5.4)a	59.2 (7.8)c	1.24	−0.7 (0.2)a

FD, fertilizer ditches; BS, bare soils. Apple production measured as fresh weight. Annual N_2O emissions and CH_4 uptake were extrapolated to the field scale using the ratio of fertilizer ditches (FD) to bare soil (BS) areas at 0.38. EF_d was calculated at the field scale. Treatment CK was not fertilized in the analyzed years. Treatment LN was fertilized at 800 kg N ha^{-1} year^{-1} in both years. Treatment HN was fertilized at 2600 and 2000 kg N ha^{-1} year^{-1} in 2010 and 2011, respectively. Values outside and inside paired-brackets are mean and standard deviation (n = 3), respectively. Values within the same column followed by the same letter do not differ at $p < 0.05$.

3.4. N_2O Fluxes

At the FD site, the seasonal variations in N_2O fluxes from the fertilized treatments were similar between the two experimental years, with large emissions from May to October (Figure 5a). The largest flux was always observed in the HN treatment, with maxima of 6.7 and 7.4 mg N m^{-2} h^{-1} in 2010 and 2011, respectively. The N_2O emissions from the CK treatment were always below 2.0 mg N m^{-2} h^{-1} during the experimental period and significantly lower than the fertilized treatments ($p < 0.01$). The difference in N_2O emissions between the LN and HN treatments was not significant ($p > 0.05$).

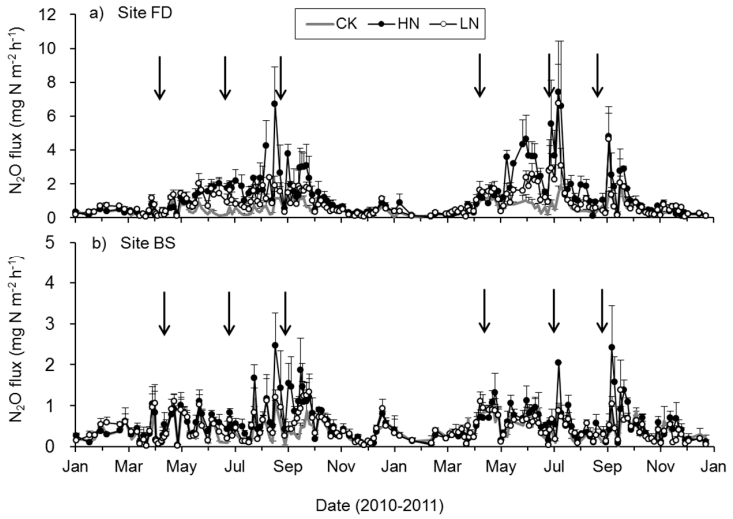

Figure 5. Seasonal variations in N_2O flux across the sampling sites and treatments during 2010–2011. (**a**) Site FD, (**b**) Site BS. FD, fertilized ditch. BS, bare soil with none ferltilizers. Data points indicate the mean of the three replicates; error bars indicate the standard error (SE). Black arrows indicate nitrogen fertilizer applications.

At the BS site, the seasonal variations in N_2O fluxes were similar between the treatments during both experimental years (Figure 5b). The largest flux was 2.5 mg N m^{-2} h^{-1}, which occurred in the HN treatment. The N_2O emissions from the HN treatment were significantly higher than the CK treatments ($p < 0.01$) in 2010. The differences of N_2O emissions between various treatments were not significant in 2011 ($p > 0.05$).

3.5. Annual N_2O Emissions and EF_d

At the FD site, annual N_2O emissions ranged from 39.1 to 112.7 kg N ha^{-1} $year^{-1}$ across the treatments and experimental years (Table 3). The annual emissions from the HN treatment were significantly higher than the CK treatment by 140% ($p < 0.01$) and 161% ($p < 0.01$) in 2010 and 2011, respectively (Table 3). The annual emissions from the LN treatment tended to be higher than the CK treatment by 75% ($p = 0.09$) and 79% ($p = 0.07$) in 2010 and 2011, respectively.

At the BS site, annual N_2O emissions ranged from 30.9 to 47.6 kg N ha^{-1} $year^{-1}$ across the treatments and experimental years (Table 3). The annual emissions from the fertilized treatments tended to be higher than the CK treatment by 9–48%. However, the differences were not significant except for the difference between the HN and CK treatments in 2010 ($p < 0.01$).

At the field scale, annual N_2O emissions ranged from 34.1 to 60.3 kg N ha^{-1} $year^{-1}$ ($n = 6$, CV = 24%) over the two experimental years (Table 3). The annual N_2O emissions from the HN treatment were significantly higher than the CK treatment by 73–77% ($p < 0.01$) in the experimental years. The annual N_2O emissions from the LN treatment were significantly higher than the CK treatment by 33–39% ($p < 0.05$) in the experimental years. The difference in annual emissions between the HN and LN treatments was also significant ($p < 0.05$). Based on these estimates, the site-specific EF_d ranged from 1.00% to 1.65% with a mean of 1.34% ($n = 4$, CV = 21%).

3.6. CH_4 Uptake

Soil CH_4 fluxes ranged from −0.53 to 0.89 mg C m^{-2} h^{-1} during the experimental period (Figure 6). The seasonal variations were quite similar between treatments and sampling sites in each year. Annual CH_4 uptake ranged from 0.4 to 1.1 kg C ha^{-1} $year^{-1}$ with non-significant differences across treatments and experimental years (Table 3).

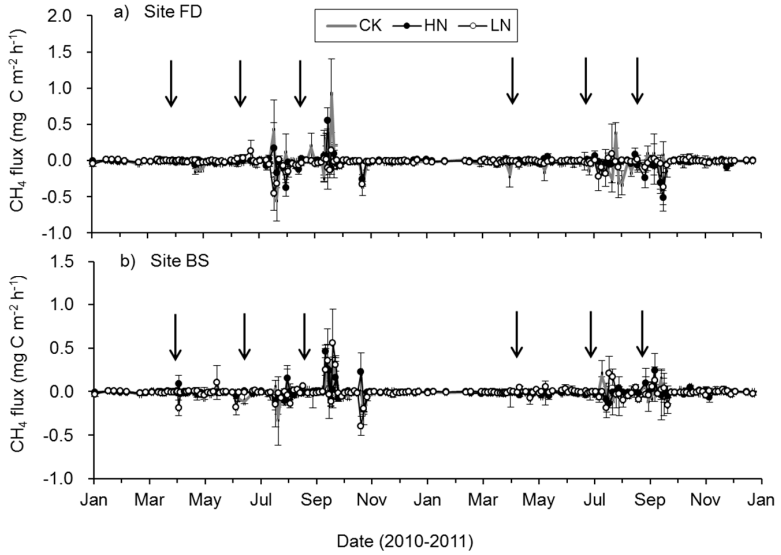

Figure 6. Seasonal variations in CH_4 flux across the sampling sites and treatments during 2010–2011. FD, fertilized ditch. BS, bare soil with none ferltilizers. (**a**) Fertilizer ditches; (**b**) Bare soil; Data points indicate the mean of the three replicates; error bars indicate the standard error (SE). Black arrows indicate nitrogen fertilizer applications.

4. Discussion

We captured both soil CH_4 production and consumption across the treatments over two experimental years (Figure 6). The net annual CH_4 fluxes indicated that all treatments were slight sinks (Table 3), which was similar to previous results obtained for various upland agricultural ecosystems [23]. Alsina et al. [34] reported that an almond orchard's soil was a minor source of CH_4, mostly due to the effect of fertigation. However, the quantities of the sink or source in those studies were negligible compared to N_2O emissions in terms of global warming potential. Our results show that the effect of N input rates on annual CH_4 uptake is not significant (Table 3).

The fact is that large N_2O fluxes, generally followed the application of N, irrigation, and precipitation, have been widely reported for various agroecosystems previously [10,34,41] and were also observed in the present study (Figure 5). We observed that the seasonal variations in N_2O fluxes related significantly to soil temperature, WFPS, DOC, NH_4^+, and NO_3^- content with small R^2 values (ranging from 0.05 to 0.2, $p < 0.01$). Given that soil WFPS was above 60% during two-thirds of the experimental period (Figure 1b), denitrification was most likely the primary N_2O source [7,8]. Soil DOC content averaged 121.6 to 169.6 mg C kg^{-1} across the sampling sites and related significantly ($R^2 = 0.84$, $n = 6$, $p = 0.01$) to annual N_2O emissions at the site scale (Figure 7a). Soil NH_4^+ and NO_3^- content averaged 121.6 to 169.6 5.0 to 103.4 mg N kg^{-1} and 16.8 to 73.6 mg N kg^{-1} across the sampling sites, respectively, and related significantly ($R^2 > 0.79$, $n = 6$, $p < 0.02$) to annual N_2O emissions at the site scale (Figure 7). The results indicated that the ability of C and N substrates regulated long-term N_2O emissions. Additionally, annual N_2O emissions at the field scale exhibited positive correlations with N input rates across the two experimental years ($R^2 = 0.95$, $n = 6$, $p < 0.01$).

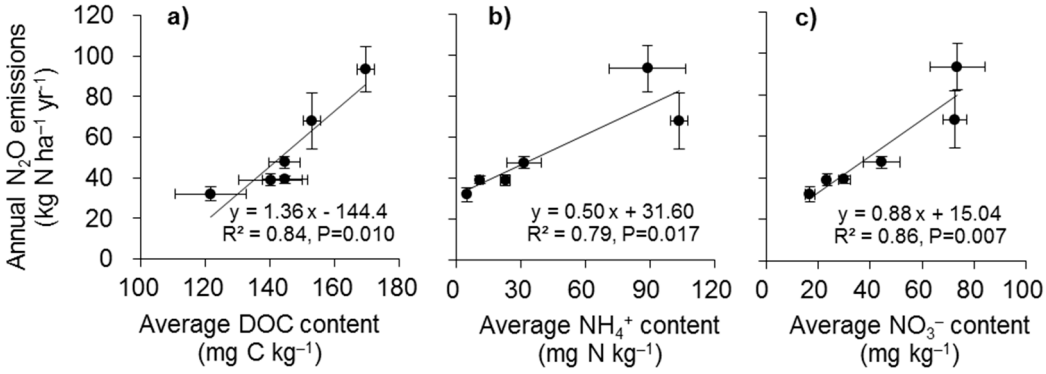

Figure 7. Relationships between annual N_2O emissions (kg N ha^{-1}) and (**a**) annual mean DOC content (mg C kg^{-1}); (**b**) annual mean NH_4^+ content (mg N kg^{-1}); and (**c**) annual mean NO_3^- content (mg N kg^{-1}) at the site scale in 2010. Error bars indicate the standard error (SE).

Our measurements show that annual N_2O emissions from the fertilized treatments ranged from 45.7 to 60.3 kg N ha^{-1} $year^{-1}$ (Table 3), which is substantially greater than previous observations in upland orchards (ranging from 0.6 to 8.6 kg N ha^{-1} $year^{-1}$; [31,32,34]). Large emissions from mineral soils have also been reported up to 66.6 kg N ha^{-1} $year^{-1}$ [41,42] based on crop type (cereal and vegetable) and N input rates (ranging from 1100 to 1700 kg N ha^{-1} $year^{-1}$, mostly in the form of dairy manure and sewage sludge). Chang et al. [42] and Mei et al. [41] reported that the annual emissions from unfertilized treatments were relatively small (0.5 to 4.1 kg N ha^{-1} $year^{-1}$); therefore, the majority (92% to 99%) of the N_2O emissions from fertilized treatments originated from the application of N fertilizer in the analyzed year. These results were reasonable because organic manure imported large N and C substrate quantities and favored microbial activities. However, in our study the large N_2O losses did not appear to be caused by N fertilization in the studied year. Our results show that the fertilizer-induced N_2O emissions contributed only 25–44% to annual losses, with background emissions accounting for the majority of emissions (ranging from 34.1 to 34.3 kg N ha^{-1} $year^{-1}$;

Table 3). Generally, background N_2O emissions from mineral soils are limited by the availability of N substrates and originate from N sources such as the decomposition of soil organic matter, atmospheric deposition, and N substrates remaining within the soil profile due to N inputs in previous years or seasons [15,43]. Globally, background emissions from mineral soils occur at a mean 1 kg N ha^{-1} $year^{-1}$ (ranging from −0.6 to 3.2 kg N ha^{-1} $year^{-1}$; [15]). Gu et al. [43,44] reported that background emissions from cropland averaged 1.21 kg N ha^{-1} $year^{-1}$ (ranging from 0.1 to 2.63 kg N ha^{-1} $year^{-1}$) and responded to SOC content (ranging from 4.5 to 50.3 g C kg^{-1}). However, this does not explain the results in our study because the site-specific SOC content was only 10.6 g C kg^{-1} (Table 1). Instead, the background emissions were most likely due to NO_3^- accumulation in the soil profile resulting from the overuse of N fertilizer throughout the area's history. Ju et al. [25] reported that the NO_3^- stored within the top 1 m of soil profiles in the study region averaged 600 kg N ha^{-1} in typical winter wheat-maize crop rotation fields and up to 1410 kg N ha^{-1} in some lands cultivated with apple trees and vegetables after the long-term overuse of N fertilizer. These NO_3^- stocks may contribute to background emissions when they are denitrified. Jahangir et al. [45] (2012) reported that denitrification potential in subsoils (deep to 1.2–1.3 m) under temperate grassland was stimulated by 450–830%, given adequate C sources. In the present study, the tree roots likely readily provided C substrates to feed microbes within the orchard's soil profile. In addition, irrigation may also introduce significant NO_3^- substrates (approximately 100–200 kg N ha^{-1} $year^{-1}$), particularly given that the NO_3^- content in the underground water averaged approximately 100 mg N L^{-1}.

In the present study, although N_2O emissions from the orchard were very high, the site-specific EF_d (averaged 1.34%) was similar to the IPCC default of 1% [19]. The results suggested we can use the IPCC default of 1% to estimate N_2O emissions for the fertilized orchards. N input rates average 842 kg N ha^{-1} $year^{-1}$ in apple orchards [27], and the apple orchard area covered 0.30 Mha in Shandong Province in 2015 [46]. Regional direct N_2O emissions from orchards were estimated at 3.4 Gg N $year^{-1}$. Pang et al. [32] found a smaller EF_d (0.66%) for region-specific apple orchards in Northwestern China and estimated the regional direct N_2O emissions at 2.5 Gg N $year^{-1}$. Therefore, the total direct emissions from apple orchards in the two regions were approximately 5.9 Gg N $year^{-1}$, which accounts for 2.1% of the total direct N_2O emissions from cropland in China [18]. That said, background emissions were not included in those regional assessments and could be much larger (such as 4–5 times in our study, assuming an N input rate of 842 kg N ha^{-1} $year^{-1}$) than the direct emissions. Reducing background emissions rather than direct emissions appears critical to the mitigation of regional N_2O emissions. More background emission data are therefore needed in the study region.

Our results indicate that high rates of N fertilization led to increasing N_2O losses without economic gains in apple yields (Table 3). Although apple production in the fertilized treatment areas was 14–32% larger than that in the unfertilized treatment areas during the two experimental years, the differences were mostly not significant (Table 3). Given that the N content within a fresh apple is approximately 0.6% (unpublished data in this study), approximately 23–27% and 7–9% of the N fertilizer was recovered in the apple harvest from the LN and HN treatments, respectively. Taking into account the production in the unfertilized treatments, we estimated that approximately 94–99% of the N input in the analyzed year was lost to the environment. Overuse of N fertilizer in China has resulted in serious environmental issues such as NO_3^- leaching, soil acidification, and greenhouse gas emissions [27,30,41,47]. The framework of "integrated nutrient management" was recently developed with incredible potential to simultaneously increase crop yields and nutrient use efficiency [28,48]. We strongly advise that conventional fertilization regimes be avoided or reduced and a much lower N input rate is recommended in the study region. The environmental and economic benefits (such as reducing N_2O emissions and increasing apple yields) of reducing N input rates may be not significant (Table 3), but economic savings (such as reduced N fertilizer costs) are certain. Still, much work is needed to determine the fate of N substrates stored in the soil profile as a result of historical fertilization.

5. Conclusions

We measured N_2O and CH_4 fluxes regularly between 2010 and 2011 in an irrigated apple orchard in Eastern China. The soil was a significant N_2O source and a minor CH_4 sink. The contribution of fertilizer-induced N_2O emissions to annual losses was moderate, resulting in an EF_d of 1.34%. Large background emissions were most likely related to historically accumulated NO_3^- in the soil profile because of the overuse of N fertilizer in past decades. Further studies on the fate of NO_3^- accumulation are urgent because reducing background emissions appears to be critical to the mitigation of regional N_2O emissions. Considering that high rates of N fertilization have led to increasing N_2O losses without economic gains in apple yields, we strongly advise that the conventional fertilization regimes be avoided, and reduced N input rates must be recommended in the study region.

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