

Depth-dependent greenhouse gas production and consumption in an upland cropping system in northern China

Yuying Wang^{a,1}, Xiaoxin Li^{a,1}, Wenxu Dong^a, Dianming Wu^b, Chunsheng Hu^{a,*}, Yuming Zhang^a, Yiqi Luo^c

^a Key Laboratory of Agricultural Water Resources, Center for Agricultural Resources Research, Institute of Genetics and Developmental Biology, Chinese Academy of Sciences, Shijiazhuang 050021, China

^b Multiphase Chemistry Department, Max Planck Institute for Chemistry, Mainz 55128, Germany

^c Department of Microbiology and Plant Biology, University of Oklahoma, Norman, OK, USA

ARTICLE INFO

Handling Editor: Junhong Bai

Keywords:

Greenhouse gas
Surface gas exchange
Subsurface gas flux
Depth-dependent contribution
Upland cropping system

ABSTRACT

Vertical profiles of greenhouse gas (GHG) production and consumption within soils have not been carefully quantified. The objective of this study was to quantify the depth-dependent contributions of CO₂, CH₄ and N₂O fluxes in the soil profile to soil surface gas exchange. We simultaneously measured the soil surface GHG emissions and the subsurface fluxes (0–115 cm) in situ by using a static chamber-based method (CM) and a concentration gradient-based method (GM) respectively, over two-year period in a maize-based upland cropping system in northern China.

We found that unfertilized maize-based farmland acted as CO₂ sources and CH₄ and N₂O sinks. Soil surface respiration was mostly contributed by the 0–15 cm horizon; while CH₄ and N₂O consumption originated from the 0–40 and 0–15 cm soil horizons, respectively. Specifically, we revealed that the soil surface respiration was contributed by the 0–5 and 5–15 cm horizons, accounting for 70.9 and 27.3% of the surface exchange, respectively. The CH₄ consumption at 0–5, 5–15 and 15–40 cm depths accounted for 54.1, 32.3 and 12.1% of the surface exchange, respectively. And the N₂O consumption at 0–5 and 5–15 cm depths accounted for 80.4 and 6.6% of the surface exchange, respectively. The subsoil below 15 cm acted largely as a CO₂ buffer; the production/consumption potentials of CH₄ and N₂O were very weak below 40 and 15 cm depths, respectively. In conclusion, our results highlight that the topsoil (0–40 cm) plays a critical role in CO₂ production and CH₄ and N₂O consumption in an unfertilized maize-based farmland in Taihang mountain areas of northern China. However, the mechanisms responsible for changes in stored greenhouse gas within soil pore space are not clear, and further observational and experimental research is required to understand those processes.

1. Introduction

The production and consumption of greenhouse gases (GHGs) such as CO₂, CH₄ and N₂O in soils is directly related to soil processes and climatic changes (Fest et al., 2009). The last 2 decades have seen an upward trend in the number of publications referencing the closed surface chamber technique for estimation of CO₂, CH₄ and N₂O soil to air fluxes (Livingston and Hutchinson, 1995; Mosier et al., 1998; Valentini et al., 2000; Hütsch, 2001; Flessa et al., 2002; Guo and Zhou, 2007; Fest et al., 2009; Petersen et al., 2011; Banger et al., 2012; Fang et al., 2014; Sanz-Cobena et al., 2012, 2014; Burgos et al., 2015). A fair understanding of the GHG exchange between soil and atmosphere is achieved from these studies, but information regarding the depth

distribution of GHG production/consumption patterns in soil profiles is still lacking. It is mostly assumed that soil surface effluxes of CO₂, CH₄ and N₂O are instantaneous soil emissions, thereby neglecting possible changes in the volatile subsurface carbon (C) and nitrogen (N) pools. However, the quantity of GHGs in soil air and in soil solution can amount to several times those of the daily soil-surface emissions (Maier et al., 2010; Wang et al., 2014). Thus, gaining a better understanding of GHG production/consumption processes in soil profiles is an essential step to accurately predict ecosystem C and N dynamics in response to climate change. Consequently, detailed knowledge of the contributing processes of subsurface GHG fluxes to surface exchange is crucial.

The production and consumption of GHGs in soils is related to environmental factors, microbiological processes and crop management

* Corresponding author.

E-mail address: cshu@sjziam.ac.cn (C. Hu).

¹ These authors contributed equally to this work.

Table 1
Soil characteristics at the experimental site.

a) Prior to the study in 2011													
Soil depth (cm)	pH (H ₂ O)	Sand (%)	Silt (%)	Clay (%)	Textural class ^a	Dry bulk density (g cm ⁻³)	Total organic matter ^b (g kg ⁻¹)	Total nitrogen ^c (g N kg ⁻¹)	Total phosphorus ^d (g P kg ⁻¹)	Total potassium ^e (g K kg ⁻¹)	Available nitrogen ^f (mg N kg ⁻¹)	Available phosphorus ^g (mg P ₂ O ₅ kg ⁻¹)	Available potassium ^h (mg K ₂ O kg ⁻¹)
0–5	7.75	71.7	21.5	6.8	SSL	1.11	19.0	1.25	0.58	20.6	88.8	12.3	221.6
5–15	7.70	71.1	19.7	9.2	SSL	1.15	16.8	1.13	0.53	20.1	81.3	7.9	154.4
15–40	7.32	70.5	18.4	11.1	SSL	1.28	8.5	0.65	0.35	19.1	42.7	3.5	140.0
40–65	7.61	72.2	13.7	14.0	SSL	1.40	5.1	0.44	0.25	19.5	26.7	3.1	118.0
65–90	7.51	65.2	12.3	22.5	SL	1.44	5.0	0.44	0.20	18.3	25.3	3.1	103.0
90–115	7.48	61.7	11.6	26.7	SL	1.46	4.0	0.36	0.18	19.8	21.4	4.0	99.2

b) After the study in 2013																
Soil depth (cm)	pH (H ₂ O)	Sand (%)	Silt (%)	Clay (%)	Textural class	Dry bulk density (g cm ⁻³)	Total organic matter (g kg ⁻¹)	Total nitrogen (g N kg ⁻¹)	Total phosphorus (g P kg ⁻¹)	Total potassium (g K kg ⁻¹)	Available nitrogen (mg N kg ⁻¹)	Available phosphorus (mg P ₂ O ₅ kg ⁻¹)	Available potassium (mg K ₂ O kg ⁻¹)	N–NH ₄ ⁺ (mg kg ⁻¹)	N–NO ₃ ⁻ (mg kg ⁻¹)	Mineral nitrogen (kg N ha ⁻¹)
0–5	7.74	71.6	21.6	6.8	SSL	1.11	19.2	1.22	0.57	21.4	88.2	12.2	211.7	0.88	1.25	1.18
5–15	7.70	71.0	19.8	9.2	SSL	1.15	16.7	1.13	0.53	19.6	82.7	7.6	155.5	0.61	0.83	1.66
15–40	7.31	70.6	18.5	10.9	SSL	1.28	8.8	0.66	0.31	18.1	43.9	3.3	140.8	0.48	0.68	3.72
40–65	7.62	72.3	13.8	13.9	SSL	1.40	5.1	0.45	0.21	18.5	27.2	2.6	116.3	0.35	0.12	1.63
65–90	7.51	65.0	12.5	22.5	SL	1.44	4.9	0.43	0.20	17.1	24.7	3.0	102.5	0.30	0.10	1.45
90–115	7.48	61.5	11.6	26.9	SL	1.46	3.9	0.37	0.17	19.4	21.4	3.3	98.3	0.22	0.03	0.93

^a SSL, sandy silt loam; SL, sandy loam.
^b Total organic carbon was measured by potassium dichromate (K₂Cr₂O₇) oxidation at 170–180 °C followed by titration with 0.1 mol l⁻¹ ferrous sulfate. Total organic matter content is deduced by the percentage of organic carbon in soil by the ‘Van Bemmelen’ factor of 1.724.
^c Total nitrogen was assayed by the Kjeldahl method after H₂SO₄ digestion in the presence of K₂SO₄-CuSO₄-Se catalyst.
^d Total phosphorus was measured by the Mo–Sb colorimetry method.
^e Total potassium was measured by digestion of 0.25 g of soil previously treated in 5 ml of hydrofluoric acid (40%) and 1.5 ml of HClO₄ (65%) according to the AFNOR standard X31-147 followed by flame photometry measurement.
^f Available nitrogen was measured by the alkaline hydrolysis diffusion method.
^g Available phosphorus concentration was measured by using an acid-extracted molybdenum colorimetric method with a HClNH₄F digestion.
^h Available potassium content is determined by using the CH₃COONH₄ extraction method, and then followed by flame photometer measurement.
ⁱ Fresh field-moist soil samples were extracted with deionized water and also with 0.5 M KCl solution. 10.0 ± 0.01 g soil subsamples were extracted in duplicate with 50 ml of 0.5 M KCl or 50 ml of deionized water by shaking at 150 rpm for 1 h and filtering the extracts through Whatman No. 42 filter papers. The soil extracts were analyzed using a standard Bran p Luebbe Auto-Analyser-3 manifold and matrix-matched standards to find the concentrations of ammonium-N and nitrate-N.

Table 2
Crop management activities during the observation period.

Timing	Crop management activities	
	Summer-maize growing season	Fallow season
October 1, 2011 to June 1, 2012	–	Fallow
June 1, 2012	Seeding	–
August 20, 2012	Herbicide application	–
October 10, 2012	Harvest	–
October 10, 2012 to June 19, 2013	–	Fallow
June 20, 2013	Seeding	–
August 22, 2013	Herbicide application	–
October 11, 2013	Harvest	–

practices (e.g., tillage, irrigation, fertilization) (Sanz-Cobena et al., 2014). However, due to the complex interactions between GHG production, consumption and transport in soil profiles, the relationships between GHG fluxes and climatic factors are often confused by huge spatial and temporal variations in emissions (Wang et al., 2013). Therefore, it is essential to determine the role of each of these climate factors in driving subsurface gas exchange processes. Furthermore, subsurface processes exert a significant control on C and N dynamics and hence on CO₂, CH₄ and N₂O emissions from soil (Valentini et al., 2000), but only a few investigations have determined CO₂ (Tang et al., 2003; Fierer et al., 2005), CH₄ (Gebert et al., 2011) and N₂O (Reth et al., 2008; Nan et al., 2016) concentration gradients in soil profiles, and few studies have reported simultaneous measurements of all 3 gases (Wang et al., 2013, 2014). Yet, very few studies have presented results with an aim to quantify the intrinsic contributions of GHG fluxes within soil to soil-surface gas exchange by using both a static chamber-based method (CM) and a concentration gradient-based method (GM) in situ, simultaneously. The CM that is used to measure soil-surface gas emissions is based on the gas concentration measurement (collected over a certain interval of time); and flux is calculated as a result of concentration gradient over time (Livingston and Hutchinson, 1995). Whereas, the GM that is used to measure GHG production/consumption within soils is based on the measurement of transport-driving gas concentration gradient; and flux can be calculated from the gradient of gas concentration and the effective gas diffusivity of the porous medium (Maier and Schack-Kirchner, 2014). Given this, we assume that under the same management practices, climatic and soil conditions, the soil surface GHG fluxes measured by the CM would be equal to those fluxes in the soil profile simultaneously measured in situ by the GM. Since the surface gas exchange measured by the CM can provide an integrative estimate of net gas production/consumption in soil, we then hypothesize that the simultaneous in situ measurement of gas concentrations in the soil profile and the calculation of those fluxes by using the GM may decipher the intrinsic contributions of profile fluxes to surface gas exchange. However, there is still a lack of experimental evidence to support this assumption. Moreover, the large-scale overexploitation for farming is currently taking place in Taihang mountain areas of northern China. Yet because of the climatic and geographical restrictions, the rain-fed and low N farmland accounts for over 85% of the total arable area in this region (Zhao et al., 2014). Furthermore, numerous studies have emphasized that aerated and low-fertility soils act as net CH₄ sink (Stuedler et al., 1989; Castro et al., 1994; Nakano et al., 2004; Hou et al., 2012; Fang et al., 2014). Meanwhile, based on the laboratory experiments of low-fertility soils, N₂O consumption by low N and rain-fed agro system has rarely been reported in the Taihang mountain region (Wu et al., 2013). However, direct evidence to show that these uptakes are happening or not is still a major challenge.

In order to test the hypotheses, we simultaneously measured the effluxes of soil surface CO₂, CH₄ and N₂O by using the CM, and the subsurface fluxes (0–115 cm) by using the GM in situ, from March 2012

to October 2013 in an unfertilized upland cropping system in Taihang mountain area in North China. The objectives of this study were (1) to characterize the spatial distributions of CO₂, CH₄ and N₂O concentrations in the soil profile, (2) to quantify the depth-dependent contributions of CO₂, CH₄ and N₂O fluxes in the soil profile to soil surface gas exchange, and (3) to evaluate the influence of environmental factors on GHG fluxes in the soil profile.

2. Materials and methods

2.1. Site description

The study was conducted at the Taihang Mountain Ecosystem Experimental Station (37°53'N, 114°16'E) of the Chinese Academy of Sciences at an elevation range of 247 to 1040 m, with slopes in the catchment ranging from 20 to 45°. The station is in the low mountain hilly area of the middle Taihang mountains, on the North China Plain. The climate is generally dry from October to May and rainy from June to September. The mean annual temperature is 13.0 °C and the mean annual precipitation is 560 mm of which about 77% typically occurs from June to September. The soil is a sandy loam and classified as an entisol (Soil Survey Staff, 2006). Soil characteristics of the experimental field are listed in Table 1.

2.2. Experimental set up

Field measurements were conducted during the growing season, from March to October in 2012 and 2013, in an unfertilized and rainfed farmland planted with maize (*Zea mays* L.). The farmland has been in a rotation with summer-maize and fallow and has not been fertilized since 1986. Details on crop husbandry are presented in Table 2.

2.3. Gas flux measurements

Fluxes of soil surface CO₂, CH₄ and N₂O were determined twice a month by using a closed static chamber-based method (CM), with 4 replicates. Each chamber (30 cm × 20 cm × 30 cm) was placed on a fixed framework between the maize rows, which was inserted approximately 5 cm into the soil with the above ground rim at about 5 cm height. Chambers were placed on frameworks using a water seal to prevent leakage during measurement. Gas samples were taken in the field using 60 ml syringes (Dalian Delin Gas Packing Co., Ltd., China) between 9:00 A.M. and 11:00 A.M. at 0, 10, 20 and 30 min after initiating the measurement.

Fluxes of CO₂, CH₄ and N₂O in the soil profile were simultaneously determined in situ by using a concentration gradient-based method (GM), with 4 replicates, following the procedure described by Wang et al. (2013). Subsurface soil gas tubes were installed at depths of 5, 15, 40, 65, 90 and 115 cm, respectively (Fig. S1). These samplers were constructed from polyvinylchloride tubes with a 2.5 cm inner diameter. The lower part of each sampler was perforated and covered by a water impermeable membrane, which only allowed soil air to diffuse into the sampler from the surrounding soil. The samplers were connected to the surface by microbore polytetrafluoroethylene (PTFE) tubing (inner diameter 0.25 cm; outer diameter 0.30 cm) fitted with three-way stopcocks (Fig. S1). The samplers were carefully inserted in the pre-drilled holes, made by a 3.0 cm diameter hand auger. The spaces around the tubes were backfilled with the soil in the same order as it was dug out. Gas sampling started after 7 months of the installation of these samplers. Gas samples were taken using the same plastic syringes, with the attached three-way stopcocks connected to the tubes via PTFE tubing at the soil surface (Fig. S1). Four samples of ambient air (0 cm) were collected as well.

The gas samples were analyzed within 24 h of collection by a gas chromatograph (Agilent GC-6820, Agilent Technologies Inc., Santa Clara, CA, USA) with separate flame ionization and electron capture

detectors (FID at 200 °C and ECD at 330 °C) for CO₂ (or CH₄) and N₂O, respectively.

2.4. Environmental factor measurements

Alongside the measurements of soil surface GHG fluxes, soil temperature was measured using geothermometers (liquid-in-glass), which were installed at 3 cm soil depth. Soil water content (0–3 cm) was determined by weighting samples before and after drying at 105 ± 0.5 °C. In the soil profile, soil temperature was measured by using 6 CS107b soil temperature probes (Cambell Scientific Inc., Logan, UT), which were installed at depths of 5, 15, 40, 65, 90 and 115 cm, respectively. A neutron access tube (120 cm length) was installed in each plot, and the soil water content of the profile was measured using a neutron moisture meter. Daily rainfall, air temperature and barometric pressure were recorded at a weather station at the experimental site.

2.5. Soil analyses

Soil core samples were collected from different soil depths (0–5, 5–15, 15–40, 40–65, 65–90, 90–115 cm) in the farmland in October 2011 (prior to the study) and October 2013 (after the summer maize harvest). Three different sub-samples, taken from a cross-section around the soil auger, were mixed to make a specific representative soil sample for each depth from each point. The samples were sealed in dark plastic bags immediately after sampling and stored at 4 °C until NO₃-N and NH₄-N determinations. The rest of the soil samples were air-dried and ground to pass a 5-mm sieve, and visible roots were removed before analysis. For measurement of soil chemical characters, the soil was further ground to pass a 0.5-mm sieve.

Soil pH was measured in a suspension of 5 g soil with 25 ml distilled water, 1 h following shaking. Soil particle size analysis was performed using the Bouyoucos Hydrometer method (Bouyoucos, 1936). Soil bulk density was determined by using the cutting ring method (Brasher et al., 1966). And all analyses of soil chemical properties were based on the standard methods for soil analyses described by Sparks (1996). Results were expressed in terms of 105 °C oven-dried weight.

2.6. Calculation of GHG fluxes

2.6.1. Soil surface GHG fluxes

Soil surface fluxes of CO₂, CH₄ and N₂O were calculated using the following equation by Hutchinson and Livingston (1993):

$$F = 60 \times 10^{-5} \left(\frac{T_0}{T_0 + T} \right) \frac{P}{P_0} \rho H \frac{dC}{dt} \quad (1)$$

Here, F is the flux rate of GHG (mg m⁻² h⁻¹); ρ is the gas density at T_0 (273.15 K) and P_0 (101.3 kPa), T is the air temperature inside the chamber (°C), P is the air pressure of the experimental site (kPa), H is the headspace height of the chamber (cm), t is the time for sampling (min), C is the gas mixing ratio by volume (10⁻⁹), dC/dt is the increase rate of gas concentration in the closed chamber (10⁻⁹ min⁻¹).

Cumulative CO₂, CH₄ and N₂O fluxes at the soil surface were estimated from linear interpolation: we multiplied the mean values of CO₂, CH₄ and N₂O fluxes of 2 consecutive sampling dates by the interval between sampling days and then calculated the sums over a time period (Dobbie et al., 1999; Chatskikh and Olesen, 2007).

2.6.2. Subsoil GHG fluxes

Subsoil fluxes of CO₂, CH₄ and N₂O can be estimated on the basis of soil gas concentration gradients using Fick's 1st law:

$$q = -D_p \frac{\delta C}{\delta z} \quad (2)$$

Here q is the gas flux density (g gas m⁻² soil s⁻¹), D_p is the soil-gas diffusivity (m³ soil air m⁻¹ soil s⁻¹), C is the gas concentration in the air-filled pore space (μmol m⁻³), z is the distance between two soil layers (m), $\delta C/\delta z$ is the vertical soil gas gradient (g gas m⁻³ soil air m⁻¹ soil).

D_p can be estimated as

$$D_p = \xi D_a \quad (3)$$

where ξ is the gas tortuosity factor, and D_a is the gas diffusion coefficient in the free air (m² air s⁻¹).

The variation of D_a with temperature and pressure is given by

$$D_a = D_{a0} \left(\frac{T}{T_0} \right)^\alpha \left(\frac{P_0}{P} \right) \quad (4)$$

where T is the air temperature (K), P is the air pressure (kPa). D_{a0} a reference value of D_a at T_0 (273.15 K) and P_0 (101.3 kPa), and is given as 0.139 cm² s⁻¹, 0.188 cm² s⁻¹, and 0.143 cm² s⁻¹ for CO₂, CH₄ and N₂O, respectively (Lerman, 1979; Pritchard and Currie, 1982). The exponent α is 1.79 for CO₂ and N₂O, and 1.82 for CH₄ (Pritchard and Currie, 1982; Mast et al., 1998).

There are several empirical models used to determine ξ (Sallam et al., 1984). We used the Millington-Quirk model (Millington and Quirk, 1961):

$$\xi = \varepsilon^{10/3}/E^2 \quad E = 1 - \frac{\rho_b}{\rho_s} \quad \varepsilon = E - \theta \quad (5)$$

where ε is the soil air filled porosity (m³ air m⁻³ soil), and E is the soil porosity (m³ voids m⁻³ soil), ρ_b is the dry bulk density (g m⁻³) (Table 1), ρ_s is the average bulk density of surface soil (2.65 g m⁻³); θ is the volumetric soil water content (VWC, %), which was measured by using a neutron moisture meter.

Note that the rate of gas production/consumption in the soil profile obtained in this study had the same unit of soil surface flux. Cumulative gas fluxes in the soil horizons were calculated following the same method described above.

2.7. Statistical analyses

Statistical analyses were performed using the SPSS software package for Windows (Version 13.0, SPSS Inc., Chicago, USA). The significance of differences in GHG concentrations and fluxes at the soil surface and in the soil horizons was tested by one-way ANOVA followed by Fisher's tests, and the standard error (SE) and least significant difference (LSD) were calculated for comparison of means. The association between gas concentration and soil gradient was evaluated by exponential regression on data averaged among replicates in the dry and wet seasons, respectively. Simple linear regression was used to evaluate the relationships between environmental variables and GHG concentrations and fluxes separately. The differences were only considered significant when $p < .05$. The Origin 8.0 software package (Origin Lab Ltd., Guangzhou, China) was used for the graphical outputs.

3. Results

3.1. Environmental conditions

Strong seasonal patterns in daily mean air temperature were clearly evident, with monthly means ranging from -9.6 (December 2012) to 30.5 °C (August 2013) and annual means of 12.3 and 12.7 °C in 2012 and 2013, respectively (Fig. 1a). Strong seasonal patterns in soil temperature were apparent at all gradients and mostly followed the air temperature changes. At the soil surface, the soil temperature at 3 cm depth ranged from 7.8 to 35.6 °C; in the soil profile, the soil temperatures across all 6 layers ranged from 5.5 to 33.7 °C (Fig. 1b). The total annual precipitation over the 2 years ranged from 418.7 to 616.5 mm,

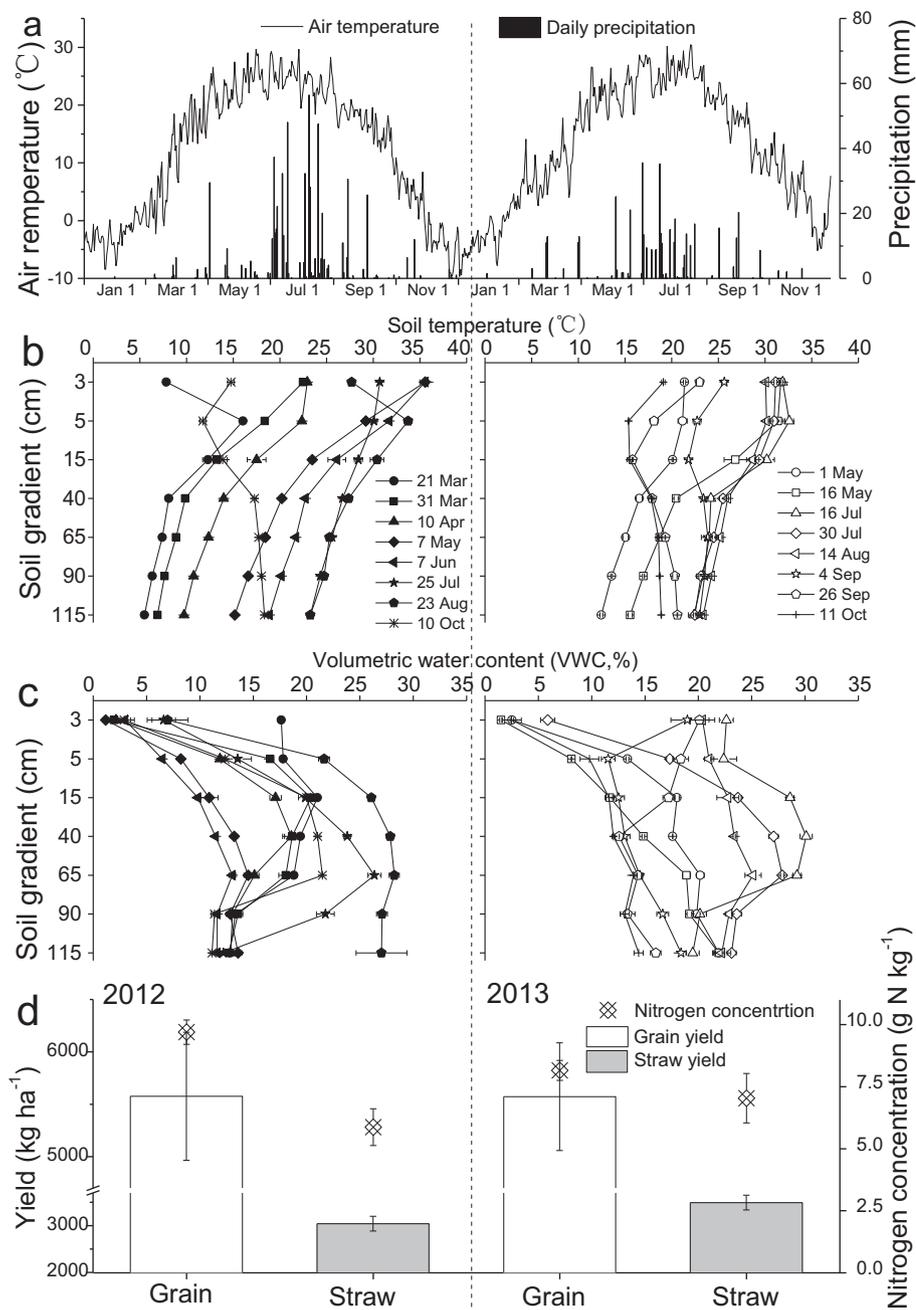


Fig. 1. Daily mean air temperature and daily sum of precipitation during the entire 2012 and 2013 period (a); soil temperature (b) and soil volumetric water content (c) at soil surface (0–3 cm) and various soil gradients (5–115 cm) during the observational period; maize grain and straw yields and their nitrogen concentrations (d). The legend is the same for graphs b and c for each year. Error bars represent standard error (n = 4).

Table 3
Average concentrations and flux rates of CO₂, CH₄ and N₂O within the soil profile.

Soil gradient (cm)	Concentration			Soil horizon (cm)	Flux rate		
	CO ₂ (ppmv)	CH ₄ (ppmv)	N ₂ O (ppbv)		CO ₂ (mg CO ₂ m ⁻² h ⁻¹)	CH ₄ (μg CH ₄ m ⁻² h ⁻¹)	N ₂ O (μg N ₂ O m ⁻² h ⁻¹)
0	483 (10.1)g	2.09 (0.008)a	566 (5.69)d	Soil surface	421 (7.72)a	- 106 (9.27)e	- 57.3 (1.27)e
5	1536 (25.6)f	1.71 (0.060)b	471 (2.24)e	0–5	315 (1.43)b	- 52.4 (3.34)d	- 42.0 (0.958)d
15	2813 (74.4)e	1.18 (0.108)c	465 (0.95)e	5–15	143 (2.24)c	- 35.9 (3.34)c	- 3.22 (0.221)c
40	6116 (52.5)d	0.436 (0.012)d	606 (13.9)c	15–40	120 (2.81)d	- 16.7 (2.51)b	5.47 (0.533)a
65	8016 (98.9)c	0.437 (0.021)d	654 (6.87)b	40–65	45.1 (1.04)e	- 0.112 (0.062)a	0.888 (0.164)b
90	9167 (59.9)b	0.379 (0.012)d	709 (18.3)a	65–90	43.8 (0.635)e	- 1.01 (0.148)a	1.58 (0.364)b
115	9559 (91.9)a	0.404 (0.015)d	702 (7.63)a	90–115	23.7 (1.39)f	- 0.100 (0.044)a	0.911 (0.101)b

Values (mean with coefficient of variance in the brackets, %) within the same column followed by the same letter are not significantly different as determined by ANOVA and Fisher protected LSD (*p* < .05).

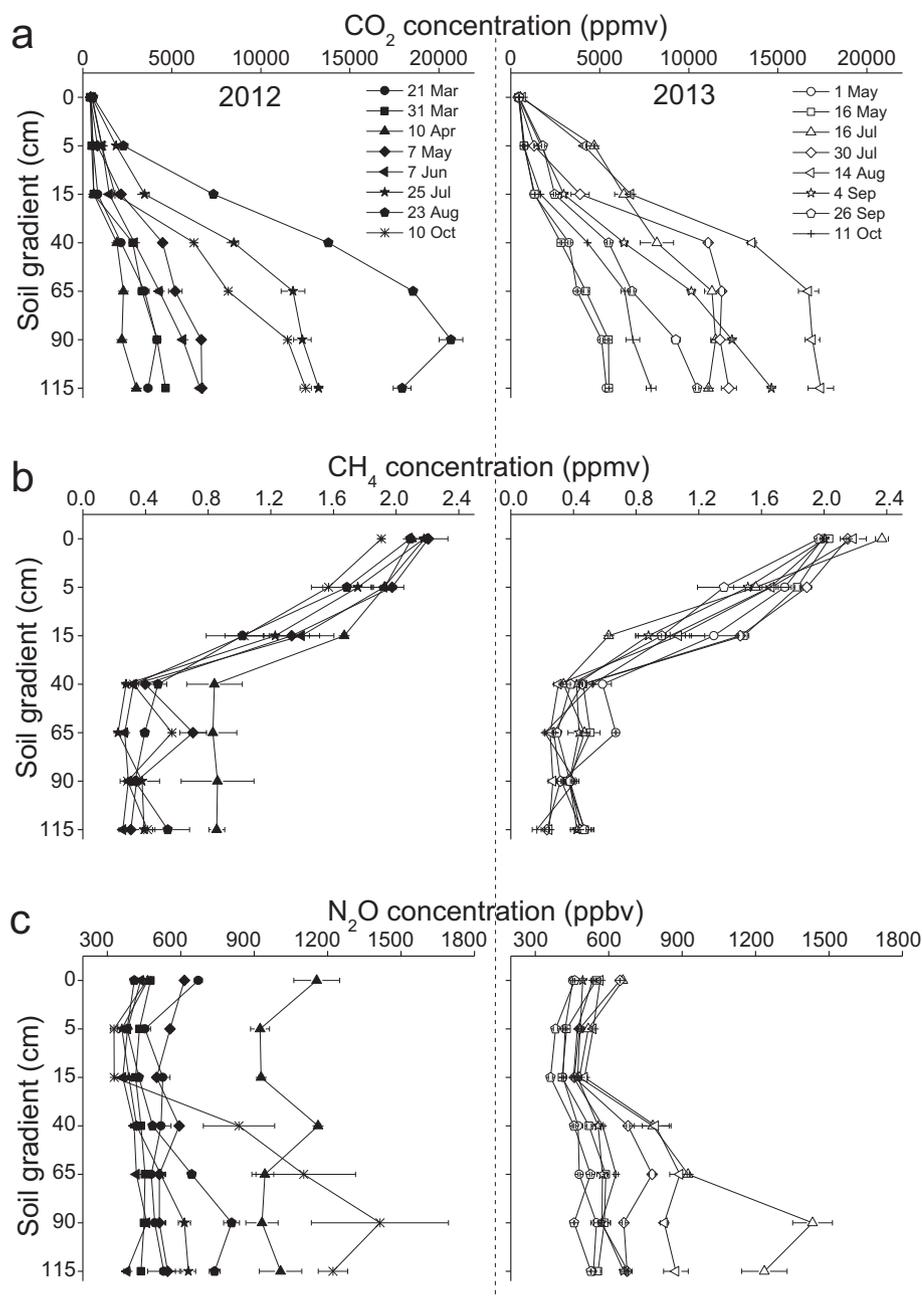


Fig. 2. Concentrations of CO₂ (a), CH₄ (b) and N₂O (c) in air (0 cm) and soil air at various soil gradients (5–115 cm) during the observational period. The legend is the same for all graphs for each year. Error bars represent standard error (n = 4).

76–84% of which fell between June and September (Fig. 1a). Soil water content (volumetric water content, VWC, %) varied with rainfall events. At the soil surface, the soil water content above 3 cm depth ranged from 1.2 to 22.6%; in the soil profile, the soil water contents across all 6 layers ranged from 6.5 to 30.1% (Fig. 1c). The grain and straw yields of maize ranged from 5573 to 5577 and 3042 to 3093 kg ha⁻¹, respectively from 2012 to 2013, and the nitrogen concentrations in grain and straw ranged from 8.2 to 9.7 and 5.9 to 7.0 g N kg⁻¹, respectively from 2012 to 2013 (Fig. 1d). The soil mineral nitrogen contents (NH₄⁺ and NO₃⁻) were 1.18, 1.66, 3.72, 1.63, 1.45 and 0.93 kg N ha⁻¹ at 0–5, 5–15, 15–40, 40–65, 65–90 and 90–115 cm layers, respectively (Table 1b).

3.2. CO₂, CH₄ and N₂O concentration profiles

Throughout the observation period, soil air CO₂ concentration increased with profile depth, and the CO₂ concentrations across all 7

layers showed statistically significant differences ($p < .05$; Table 3 and Fig. 2a). The seasonal trend in soil air CO₂ concentration was very distinct. The profile CO₂ concentrations were much higher in the wet and warm season (June to September) than in the dry and cold season (March to May) (Fig. 2a).

Throughout the observation period, soil air CH₄ concentration decreased with profile depth, and the CH₄ concentrations across the 0, 5, 15 and 40–115 cm layers showed statistically significant differences ($p < .05$; Table 3 and Fig. 2b). The seasonal trend in soil air CH₄ concentration was very weak (Fig. 2b).

Interestingly, soil air N₂O concentration decreased significantly in 0 to 15 cm soil depth, while N₂O concentration in 40 to 115 cm soil depth increased significantly (Table 3). And the N₂O concentrations across the 0, 5–15, 40, 65 and 90–115 cm layers showed statistically significant differences ($p < .05$; Table 3 and Fig. 2c). The soil air N₂O concentration peak corresponded with the low soil water content, e.g., the N₂O concentrations increased obviously with the decreases in soil water

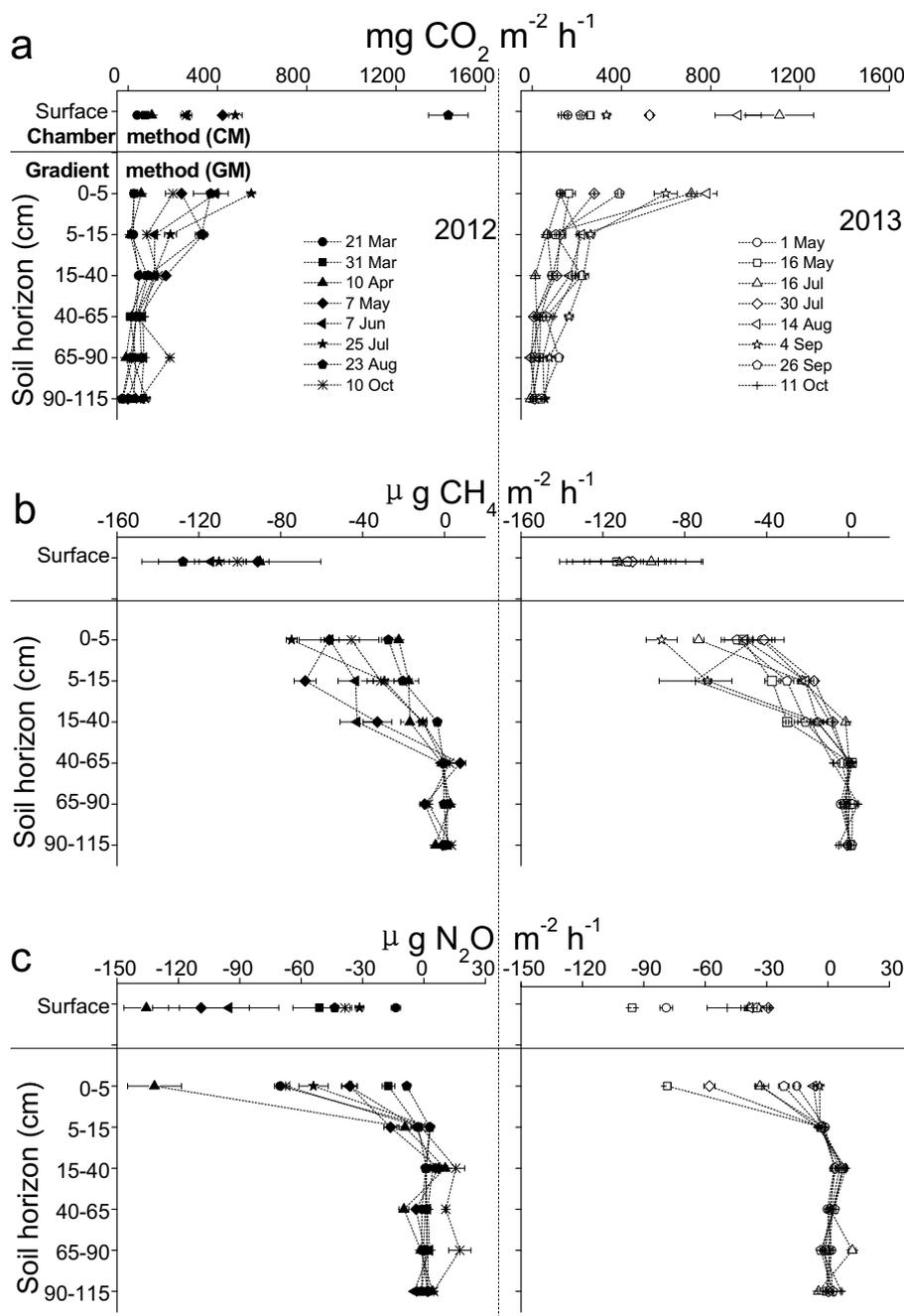


Fig. 3. Flux rates of CO₂ (a), CH₄ (b) and N₂O (c) at soil surface measured using the chamber-based method and at various soil horizons (0–115 cm) measured using the concentration gradient-based method during the observational period. The legend is the same for all graphs for each year. Error bars represent standard error (n = 4).

contents at depths > 90 cm on 10 October 2012 and 16 July 2013 (Figs. 1c and 2c). N₂O concentrations in the air and soil profile showed weak seasonal variations (Fig. 2c).

3.3. CO₂, CH₄ and N₂O fluxes

The maize-based farmland acted as a net source of CO₂ to the atmosphere based on the CM measurement of soil surface fluxes; meanwhile, the upward fluxes of CO₂ from all 6 soil horizons were found based on the GM measurement (Fig. 3a and Table 3). The CO₂ efflux rate significantly decreased with increasing soil depth ($p < .05$; Table 3). The efflux rates showed strong seasonal cycles from June to August above 15 cm depth, and remained fairly stable and low below 15 cm (Fig. 3a).

The maize-based farmland acted as a net sink for CH₄ based on the CM measurement of surface fluxes (Fig. 3b). Meanwhile, the net

influxes of atmospheric CH₄ into the top 0–5, 5–15 and 15–40 cm soil horizons were observed based on the GM measurement (Fig. 3b and Table 3), indicating that the consumption occurred in the top 0–40 cm of the soil. No obvious effects of rainfall events and growing season on CH₄ consumption rate were found at the soil surface nor in the profile, indicating that the increase in the CH₄ sink by soil moisture or plant growth was negligible. The mean CH₄ consumption rate at the soil surface was significantly higher than those at depths of 0–5, 5–15 and 15–40 cm ($p < .05$; Table 3). The CH₄ fluxes below 40 cm depth were negligible throughout the entire observational period (Fig. 3b and Table 3).

The maize-based farmland acted as a net sink for N₂O based on the CM measurement of surface fluxes (Fig. 3c). Meanwhile, the net influxes of atmospheric N₂O into the top 0–5 and 5–15 cm soil horizons were observed based on the GM measurement (Fig. 3c and Table 3), indicating the consumption occurred in the top 0–15 cm of the soil. An

Table 4
Concentrations and fluxes of CO₂, CH₄ and N₂O in response to soil depth and environmental factors.

		CO ₂ concentration			CH ₄ concentration			N ₂ O concentration		
		α	β	R ² and p	α	β	R ² and p	α	β	R ² and p
Soil depth	In dry season	535.70–950.47	0.017–0.023	0.75–0.85, < .05	1.20–1.77	–0.20 to –0.01	0.60–0.85, < .05	–	–	–
	In wet season	1281.35–2780.64	0.021–0.029	0.59–0.78, < .05	1.41–1.91	–0.02 to –0.01	0.70–0.92, < .05	386.27–551.35	0.003–0.009	0.74–0.92, < .05
Model function		Y = α × e ^{βx}								

		0–5 cm			5–15 cm			15–40 cm			40–65 cm			65–90 cm			90–115 cm					
		CO ₂	CH ₄	N ₂ O	CO ₂	CH ₄	N ₂ O	CO ₂	CH ₄	N ₂ O	CO ₂	CH ₄	N ₂ O	CO ₂	CH ₄	N ₂ O	CO ₂	CH ₄	N ₂ O			
ST	0.457	–0.047	–0.357	0.541 ^b	–0.102	0.124	0.124	0.481	0.324	0.067	0.228	0.333	–0.093	0.271	–0.030	0.310	0.025	0.172	0.155	0.281	0.393	–0.074
VWC	0.356	0.056	0.601 ^b	0.382	0.081	0.313	–0.204	–0.204	0.742 ^a	0.511 ^b	–0.572 ^b	0.824 ^a	0.331	–0.507 ^b	0.086	0.211	–0.576 ^b	0.143	–0.084	–0.559 ^b	0.228	–0.313

Y is gas concentration, X is soil depth, and α and β are best-fitting coefficients. R² is the coefficient of determination; p value is the significance probability.

ST is soil temperature (°C). VWC is volumetric water content (%).

^a Pearson's correlation coefficient, 2-tailed tests of significance.

^b Significant correlation at a < .01.

^c Significant correlation at a < .05.

increase in N₂O sink strength coincided with fairly low soil water content, e.g., the surface N₂O consumption rate increased markedly from 31 March to 7 June in 2012 during the dry spring with little rainfall (65.5 mm) and low soil moisture (1 to 2%, VWC) (Figs. 1c and 3c). The mean N₂O consumption rate at the soil surface was significantly higher than those at depths of 0–5 and 5–15 cm (p < .05), and the N₂O fluxes were negligible below 15 cm (Fig. 3c and Table 3).

3.4. CO₂, CH₄ and N₂O concentrations and fluxes in response to soil depth and environmental factors

The relationship between soil gas concentration and soil depth can be described by an exponential function (0.59 < r² < 0.92, p < .05; Table 4a). Soil air CO₂ concentration increased exponentially with profile depth, and CH₄ decreased exponentially with depth in both the dry and wet seasons (p < .05; Table 4a). We did not find an effect of soil depth on N₂O concentration in the dry season (p > .05); but N₂O concentration exponentially increased with depth in the wet season (p < .05; Table 4a).

CO₂ flux rate was significantly positively correlated with soil temperature at a depth of 0–5 cm; and the flux rates were significantly negatively correlated with soil water contents below 15 cm depth (p < .05; Table 4b). CH₄ and N₂O flux rates were significantly positively correlated with soil water contents at soil depths of 5–40 cm (p < .01) and 5–15 cm (p < .05), respectively. The soil surface N₂O flux rate was significantly positively correlated with soil water content (p < .05; Table 4b).

3.5. Cumulative fluxes of CO₂, CH₄ and N₂O

The cumulative fluxes of CO₂, CH₄ and N₂O throughout the observation period are shown in Fig. 4. The maize-based farmland acted as a net source of CO₂ to the atmosphere. The cumulative CO₂ effluxes measured by the CM at the soil surface were very close to those simultaneously measured by the GM in the top 0–15 cm soil horizon (Fig. 4a). These results suggested that the top 0–15 cm soil horizon contributed 97.9–98.8% of the cumulative CO₂ efflux at the soil surface. Thus, the soil surface respiration was contributed by the 0–5 and 5–15 cm soil horizons for 64.2–77.7 and 20.2–34.5%, respectively. Interestingly, the cumulative upward fluxes were large in the horizons below 15 cm, with values ranging from 53 to 158 and 19 to 123 g C m^{–2} in 2012 and 2013, respectively (Fig. 4a).

The maize-based farmland also acted as a net CH₄ sink. The cumulative amounts of CH₄ consumption measured by the CM at the soil surface were very close to those in the top 0–40 cm soil horizon simultaneously measured by the GM (Fig. 4b). These results suggested that the CH₄ consumption mostly occurred in the 0–40 cm horizon. Thus, the CH₄ consumption at depths of 0–5, 5–15 and 15–40 cm accounted for 51.7–56.5, 31.1–33.5 and 10.7–13.5% of the surface fluxes, respectively (Fig. 4b). The cumulative fluxes of CH₄ below 40 cm were negligible during the entire observational period.

Additionally, the maize-based farmland acted as a net N₂O sink. The cumulative amounts of N₂O consumption measured by the CM at the soil surface were –179 and –107 mg N m^{–2} in 2012 and 2013, respectively; and those in the top 0–15 cm soil horizon simultaneously measured by the GM were –161 and –90 mg N m^{–2} in 2012 and 2013, respectively (Fig. 4c). Interestingly, the amounts of soil surface N₂O consumption measured by the CM were 10.0–16.0% higher than those in the 0–15 cm horizon measured by the GM (Fig. 4c). The cumulative N₂O fluxes below 15 cm depth were negligible during the entire observational period.

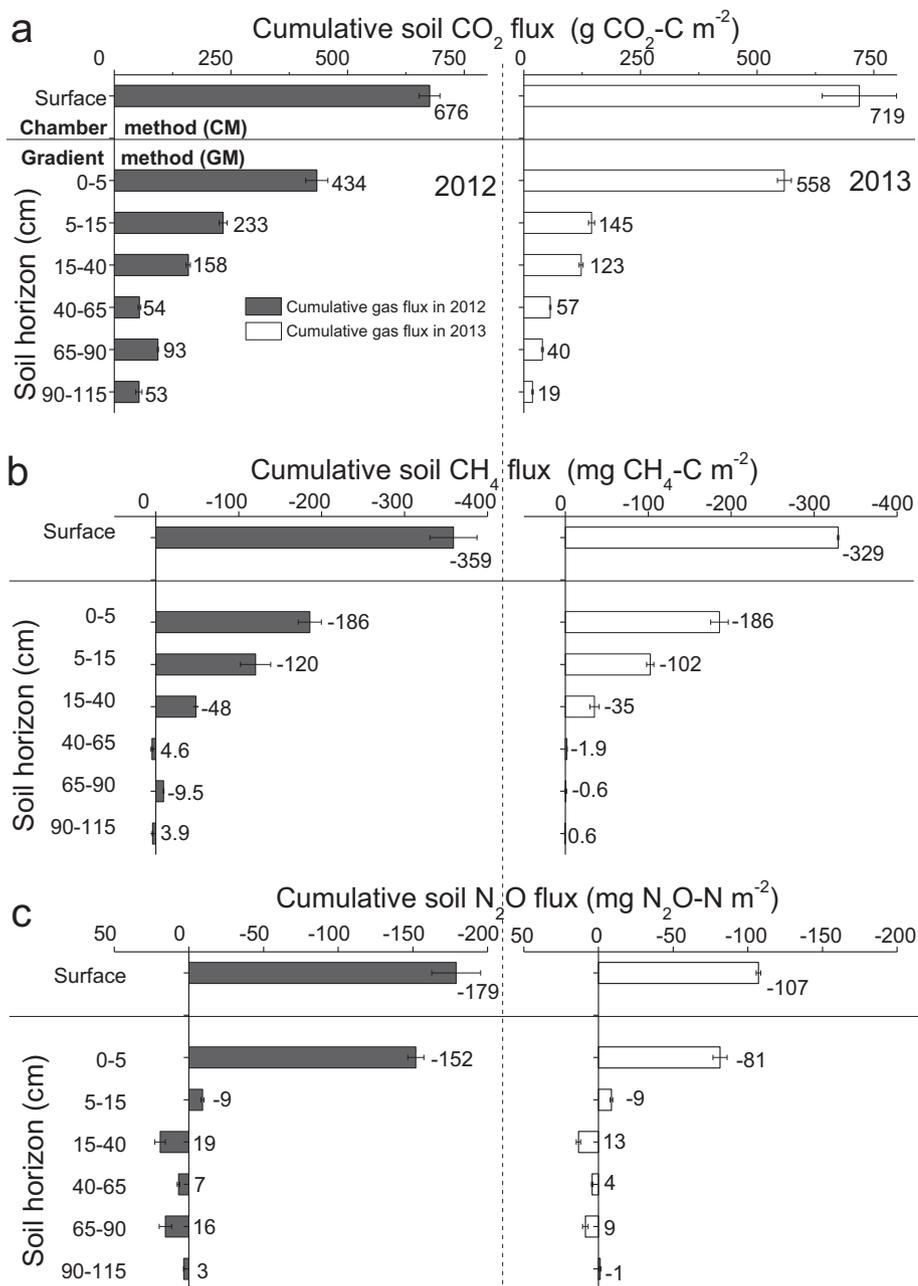


Fig. 4. Cumulative fluxes of CO₂ (a), CH₄ (b) and N₂O at soil surface measured using the chamber-based method and at various soil horizons (0–115 cm) measured using the concentration gradient-based method during the observational period. The legend is the same for all graphs for each year. Error bars represent standard error (n = 4).

4. Discussion

4.1. Soil CO₂ distribution and efflux

Soil air CO₂ concentration increased exponentially with profile depth over the 2-year field experimental period (Fig. 2a and Table 4a), which is consistent with previous findings (Fierer et al., 2005; Wang et al., 2013). CO₂ levels are often higher at the bottom of the soil profile as aeration rates are poorer at the bottom of the soil profile or root zone than in the surface soil. Higher CO₂ concentrations at greater soil depths may also be attributed to molecular weight differences among gases; those with higher molecular weights often remain at the bottom of the soil profile. The seasonal pattern of changes in CO₂ concentration in the soil profile correlated with the soil temperature envelope (Figs. 1b and 2a), and has been attributed to CO₂ respiration following the rapid growth of plant root biomass during the wet and growing season (Fierer et al., 2005; Wang et al., 2013).

Results from the 2-year field experiment showed the consistent

efflux of CO₂ by soil with a mean of 421 mg CO₂ m⁻² h⁻¹ (Fig. 3a and Table 3). The results were well within the range of published data of non-N-fertilized loam soils (from 264.3 to 446.1 mg CO₂ m⁻² h⁻¹) compiled by Mu et al. (2009) for chamber measurements on agricultural fields. The in situ CO₂ flux rates measured in our study were at the upper end of the ranges summarized from previous studies. There was very good agreement between the cumulative CO₂ efflux measured by the CM at the soil surface and those in the 0–15 cm soil horizon simultaneously measured by the GM in situ (Fig. 4a). This may suggest that the soil surface CO₂ efflux was dominated by the upward diffusion from the top 0–15 cm of the soil in the cropping system. Therefore, we conservatively estimated that the soil surface respiration was contributed by the 0–5 and 5–15 cm horizons and accounted for 70.9 and 27.3% of the exchange, respectively. Interestingly, the cumulative fluxes of CO₂ from 15 to 115 cm were fairly large, e.g., 358 and 238 g C m⁻² in 2012 and 2013, respectively (Fig. 4a). Maier et al. (2010) argue that most studies implicitly consider soil surface efflux of CO₂ as instantaneous soil respiration, thereby neglecting possible

changes in the volatile subsurface carbon pool. They found the rate of change of the stored CO₂ in the soil pore space of the vadose zone (0–100 cm) attained a significant percentage of the soil CO₂ efflux, which was > 50% in certain situations in a slow-growing 46-year-old Scots pine stand in southwest Germany (Maier et al., 2010). Maier et al. (2010) believe the quantity of CO₂ in soil air and in soil solution can amount to several times that of the daily soil respiration. Our results are in agreement with them that the soil surface respiration (CM) accounted for 65.9–76.4% of the stored CO₂ flux amounts (GM) in the soil pore space of the vadose zone (0–115 cm; Fig. 4a). We believe that the subsoil below 15 cm is neither a source nor a sink of CO₂, but more likely acts largely as a buffer.

CO₂ production in soil is strongly influenced by biological processes, such as root respiration and decomposition (Wang et al., 2014). These processes, in turn, are affected by soil temperature, water content and substrate availability. In our study, soil water content governed CO₂ flux below 15 cm in soil profile ($p < .05$; Table 4b). Our findings agree in general with most prior works (e.g., Flessa et al., 2002; Kim et al., 2007; Wang et al., 2013; Sanz-Cobena et al., 2014). However, the temperature dependences of CO₂ fluxes were not consistently strong in those horizons ($p > .05$; Table 4b). This might imply that in deeper soil profiles, soil characteristics and root growth could directly or indirectly complicate the response mechanism of CO₂ fluxes to environmental factors (Reichstein and Beer, 2008; Bond-Lamberty and Thomson, 2010). In the present study, soil chemical characteristics may play major roles in CO₂ fluxes particular in the top 0–40 cm of soil. The cumulative CO₂ fluxes in the top 0–40 cm of soil were much higher than those below 40 cm (Fig. 4a); suggesting the dependency of soil nutrient availability for CO₂ fluxes were fairly strong (Table 1). Additionally, there was a fallow period during October to June in our study (Table 2). Liebig et al. (2010) argue that carbon dioxide emission has been found to be lower in cropping systems with the inclusion of fallow due to lower carbon inputs and minimum soil disturbance. Thus, we speculated that the CO₂ flux might be lower during the fallow period than the maize growing period. But because of the lack of gas flux in fallow period in this study (October 2011 to March 2012, and October 2012 to April 2013), the contribution of fallow in feedback responses of soil respiration cannot be exported.

4.2. Soil CH₄ distribution and flux

We found a typical exponential decrease in CH₄ concentration with profile depth in the 2-year field experiment (Fig. 2b and Table 4a), which is highly consistent with previous findings (Wang et al., 2013, 2014). CH₄ concentration decreased sharply with depth in the top 0–40 cm of the soil, whereas those below 40 cm remained fairly stable (Fig. 2b), suggesting the oxidation of CH₄ by methanotrophic microorganisms mainly occurred in the 0–40 cm horizon. This corresponded fully with the results shown in Figs. 3b and 4b, that the consistent consumption of CH₄ mostly occurred above 40 cm depth. Soil CH₄ production derives from two simultaneously occurring processes, production by methanogens and consumption by methanotrophs, and the dominance of one process over another can change rapidly following small changes in soil oxygen availability (Sanz-Cobena et al., 2014). In our study, the clay content and soil bulk density below 40 cm were much higher than those in the top 0–40 cm of soil (Table 1). Consequently, it may thus be concluded that the soil O₂ concentration might be fairly higher above 40 cm than those below 40 cm (data not shown). As a consequence, CH₄ oxidation may occur mainly in the top 0–40 cm of soil. Additionally, there was a fallow period during October to June in our study (Table 2). Mosier et al. (1991) believe that fallow may enhance the capacity of the soil to act as a sink for CH₄. Thus, we speculated that the CH₄ sink might become stronger during the fallow period (dry and cold) than the maize growing period (wet and hot) due to the lower soil moistures (data not shown). However, together with the facts that O₂ concentration and fallow flux (October 2011 to March

2012, and October 2012 to April 2013) were not measured in this study, there might be small variances between these explanations and the observed data.

CH₄ consumption has been observed in nearly all types of aerated soils, such as in forests (Stuedler et al., 1989; Castro et al., 1994; Nakano et al., 2004), grasslands (Mosier et al., 1991; Fang et al., 2014), tundra (Whalen and Reeburgh, 1990) and deserts (McLain and Martens, 2006; Hou et al., 2012). However, very few published studies have examined CH₄ dynamics of soils in unfertilized upland cropping systems. Our findings illustrated the consistent consumption of CH₄ by soil with a mean of $-105.6 \mu\text{g m}^{-2} \text{h}^{-1}$ in the upland cropping system throughout the entire observation period (Fig. 3b and Table 3). Among upland soils, CH₄ is probably more efficiently oxidized in forest soils than in agricultural soils. Reported average CH₄ consumption rates by aerobic temperate-forest soils range from -373.3 to $-160.0 \mu\text{g CH}_4 \text{ m}^{-2} \text{h}^{-1}$ (Stuedler et al., 1989; Castro et al., 1994; Nakano et al., 2004). Ojima et al. (1993) reported that subjecting a soil to culture (either cultivating or planting with trees) reduced its CH₄ oxidation activity by about 60%. Similarly, we found that the CH₄ consumption rates in the unfertilized cropland soils only accounted for 28–66% of the aerobic temperate-forest soils. Furthermore, there was very good agreement between the cumulative amounts of surface CH₄ consumption measured by the CM and those in the 0–40 cm soil horizon simultaneously measured by the GM in situ (Fig. 4b). This may suggest that the CH₄ consumption occurred in the top 0–40 cm of the soil, and also offered an opportunity to quantify the relative contributions of CH₄ consumption from different soil horizons. Our findings revealed that the CH₄ consumption in the 0–5, 5–15 and 15–40 cm soil horizons accounted for 54.1, 32.3 and 12.1% of the surface fluxes, respectively (Fig. 4b). Moreover, CH₄ consumption layers in different soils are site-specific. For example, Hou et al. (2012) reported that CH₄ consumption was weak below the depth of 50 cm in a fenced sand soil, whereas our results demonstrated that the consumption was very weak below 40 cm in the sandy loam soil. This can be possibly attributed to the limitations in both atmospheric CH₄ diffusion and soil oxygen level in soils. Additionally, net CH₄ sinks were found in some plantation conditions. Lang et al. (2017) reported that the soil CH₄ uptake was stronger in rainforest than in rubber plantations and converting rainforest to rubber plantations weakened CH₄ uptake especially during the very wet period in Xishuangbanna, Southwest China. And Ruan and Robertson (2013) observed no significant difference in CH₄ uptake between a Conservation Reservation Program pasture (*Bromus inermis*) and a converted soybean (*Glycine max*) cropland in the Great Plains of Northern America. The present study indicated the unfertilized maize-based farmland acted as a net CH₄ sink in the hilly area of northern China. We speculated the differences of CH₄ uptakes in the different types of plantations may be attributed to differences in soil characteristics and soil water content.

The temperature dependence of CH₄ flux was not consistently strong ($p > .05$), and CH₄ flux and soil water content showed good agreement and consistency only in the 5–40 cm of soil ($p < .05$; Table 4b). Le Mer and Roger (2001) argue that soil temperature in less extreme environments is not always the major controlling factor in that CH₄ oxidation rates vary little over a wide range of temperatures (1–30 °C). We supposed that the general lack of sensitivity to temperature (or moisture) may partly be related to the wide diversity of methanotrophs that are typically found in soils, and partly to other interacting effects such as gas diffusion limitations.

4.3. Consumption of atmospheric N₂O by soil

We found the consistent consumption of N₂O at fairly low mean soil moisture of 9% (VWC) and high mean soil temperature of 26 °C (Figs. 1 and 3c). The mean N₂O consumption rate was $-57.3 \mu\text{g m}^{-2} \text{h}^{-1}$ (Table 3), which was close to the range of -62.9 to $-59.7 \mu\text{g N}_2\text{O m}^{-2} \text{h}^{-1}$ reported by Ye and Horwath (2016) in

rewetted peatlands converted to rice paddies during the growing season. But it was much higher than the reported $-26.4 \mu\text{g m}^{-2} \text{h}^{-1}$ in the soils across 3 high arctic polar deserts (mean soil temperature, 7°C) by Stewart et al. (2012). Chapuis-Lardy et al. (2007) argue that N_2O consumption will be significantly enhanced in soil temperatures above 5°C . Therefore, the difference can be possibly attributed to the fact that the soil temperatures in our study were much higher (26°C) than those (7°C) reported by Stewart et al. (2012). Furthermore, our results challenge the view that N_2O consumption usually takes place in ecosystems with N-limitation and high soil water content, such as fens and boreal mires (Roobroeck et al., 2010; Lohila et al., 2010), suggesting it can also occur under dry conditions in a non-N-fertilized upland cropping system. There are relatively few studies that have found N_2O sinks under dry conditions (Flechard et al., 2005; Goldberg and Gebauer, 2009; Wu et al., 2013) and the underlying mechanisms have not yet been determined (Stewart et al., 2012). One possible explanation for N_2O consumption under drier soil conditions may be due to enhanced diffusion of N_2O from the atmosphere to the site of denitrification, and in the absence of NO_3^- , N_2O may be used as the electron acceptor in denitrification, leading to N_2O consumption and N_2 formation (Chapuis-Lardy et al., 2007; Lohila et al., 2010). For example, isolated denitrifiers can grow using solely N_2O as an electron acceptor (Bazyliński et al., 1986). Furthermore, considerable evidence has been found to support the idea of an additional pathway via which many bacteria are able to respire nitrate in the presence of oxygen (Lloyd, 1993; Patureau et al., 2000). Thus, N_2O consumption by dry soil may not be controlled by anaerobic denitrification, but rather by aerobic denitrification (Wu et al., 2013). Zausig et al. (1993) reported that anaerobic centers disappeared when the aggregate pore water pressures were less than -6 kPa . That means all aggregate pore centers are oxic when soil moisture content is very low; e.g., in our study the surface soil water contents ranged from 1 to 2% (VWC) from the end of March to May in 2012 (Fig. 1c). Therefore, we may argue that the volume of anaerobic microsites in our soils might be very small, and that the N_2O consumption by dry soil in the aerobic and partly anaerobic conditions was likely not controlled by anaerobic denitrification but by aerobic denitrification. Moreover, N_2O consumption was regulated by complex interactions between O_2 concentration and soil moisture content (Wu et al., 2013). In our study, the soil bulk density and clay content were much lower in the top 0–15 cm than those below 15 cm (Table 1). Such soil characteristics are capable of regulating oxygen availability and thus favoring aerobic denitrification rates. In consequence, the soil O_2 concentration might be fairly higher in the top 0–15 cm than those below 15 cm (data not shown). Meanwhile, the soil water content in the top 0–15 cm of soil was fairly lower than those below 15 cm (Fig. 1b). Chapuis-Lardy et al. (2007) and Wu et al. (2013) believe that N_2O consumption increased significantly in dry soil with relatively high O_2 concentrations. It is therefore not surprising that we observed the N_2O consumption mainly occurred in the top 0–15 cm. In addition, low mineral N has been shown to favor N_2O consumption. The cropland in our study is a typical non N input and low-yield upland ecosystem (Table 2 and Fig. 1d), and the mineral N contents in the soil profile were fairly low (Table 1b). Such soil nitrogen nutrient conditions are favored for N_2O consumption.

Interestingly, the amounts of soil surface N_2O consumption measured by the CM were higher than those in the top 15 cm of the soil simultaneously measured by the GM in situ in both years; i.e., the disparity between the results of the 2 methods ranged from 10.0–16.0% (Fig. 4c). Such a response may imply that production, consumption and transport processes of N_2O in soils might be more complex than those of CO_2 and CH_4 . For example, Flechard et al. (2005) and Lohila et al. (2010) found that soils may be producing N_2O at the surface, but a few centimeters below, consumption of N_2O may exceed production. Furthermore, the strength of a N_2O sink is not only dependent on the potential for N_2O reduction to N_2 , but also factors such as the dissolution of N_2O into soil water within soil profile (Chapuis-Lardy et al., 2007).

Few studies have highlighted that the concentration of N_2O dissolved in soil water influences the net positive or negative fluxes measured at soil surface (Bowden and Bormann, 1986; Pihlatie et al., 2005). Meanwhile, the particle profile in the root zone contains 74% water and air pore space, which in turn can hold water and oxygen in a nearly perfect balance (Flechard et al., 2005; Lohila et al., 2010). Hence, N_2O flux may be highly influenced by soil water content in the root zone, especially in upland areas. In our study, the soil water contents in the profile were much higher than those in the top 3 cm of the soil (Fig. 1c). Consequently, we suspected that part of the N_2O , which was consumed at the active sites in the soil, might dissolve in the soil water. In an upland cropping system, water-dissolved N_2O can be removed from the soil profile in 2 ways. Firstly, emissions of water-dissolved N_2O via soil evaporation may occur. Fortunately, this part of the missing N_2O sink can be captured at the soil surface by using the CM. It means the soil evaporation might be a minimal potential factor responsible for the disparity of N_2O consumption measured by the 2 methods. Secondly, N_2O emissions via plant transpiration may occur when water-dissolved N_2O is taken up by roots and then released to the atmosphere via transpiration through plants (Chang et al., 1998; Pihlatie et al., 2005). However, unlike the GM measuring concentrations within the soil profile, the CM cannot capture this part of the missing N_2O sink because the chambers were placed between maize rows. This may suggest that plant transpiration might be the major potential factor responsible for the disparity. Our study further implied that N_2O emissions via evapotranspiration may occur when N_2O dissolves in soil water.

In our study, the relationships between environmental factors and N_2O fluxes were fairly weak (Table 4b). Several studies have demonstrated that soil N_2O fluxes are highly spatially and temporally variable, which makes it extremely difficult to determine the impact of individual driving factors (Hutchinson and Livingston, 1993; Livingston and Hutchinson, 1995; Mosier et al., 1998). We speculated that the weak correlations between N_2O fluxes and environmental factors might be attributed to relationships being hidden within this high spatial and/or temporal variation. Furthermore, net N_2O sinks were found in some plantation conditions (Ye and Horwath, 2016), but plant-mediated N_2O uptake or emission has yet to be consistently identified. Our data implied the emissions of soil water-dissolved N_2O via plant transpiration may account for 13.0% of the surface exchange. However, the measurement is limited because the plant-mediated N_2O flux was extrapolated from 2 different measurement methods. Thus, to be able to consider N_2O sinks in the global N_2O budget, more research is required to better understand the mechanism underlying plant-mediated N_2O flux. Moreover, there was a fallow period during October to June in our study (Table 2). The effects of fallow on N_2O emission are fairly complicated. Greater N_2O flux has been observed under fallow relative to cropped phases when no N is applied (Mosier et al., 1991). Whereas, Dusenbury et al. (2008) found N_2O emission during fallow was lower than during cropped phases when N is applied. Thus, we speculated that the N_2O sink might become weaker during the fallow period than the maize growing period in the typical non N input upland system (data not shown). But because of the lack of gas flux in fallow period (October 2011 to March 2012, and October 2012 to April 2013), there might be variances between our speculation and the observed data.

4.4. General considerations regarding employed methods

Although there was good consistency between the cumulative amounts of soil surface CH_4 consumption measured by the CM and those in the top 0–40 cm of the soil measured by the GM, there was still a disparity of 1.5% between them (Fig. 4). We suspect that a potential source of the bias is the sampling method of the CM. Chamber methods depend exclusively on headspace gas concentration measurements and provide an indirect measure of gas flux across the soil surface only under steady-state conditions. Thus, the challenge of using CM is to minimize perturbations by the chamber of not only the underlying rates

of root and microbial respiration, but also the transport and emission phenomena that determine what fraction of total gas production reaches the headspace of the chamber during its period of deployment (Rochette and Hutchinson, 2004). Since the chamber was placed at the soil surface at least 2 min before the measurement without the presence of collar, it could be possible that the higher gas uptake or flux in the static chamber than in the soil profile resulted from increased soil disturbance, e.g., soil/air temperature and moisture, pressure fluctuations, site disturbance, gas concentration gradient, and air mixing regime. Furthermore, although the experiment field is located in a pristine area, there is a coal mining area of Shanxi province about 150 km northwest of the station. It might affect the fluctuations of atmospheric CO₂, CH₄ and N₂O concentrations.

Ultimately, it is always difficult to extrapolate results from 2 different measurement methods, especially at the field scale. Generally, the applicability of the results depends on the measurement site, measurement setup and accuracy of the flux measurement. Based on the good consistency of CH₄ or CO₂ fluxes between the 2 methods (Fig. 4a and b), we believe that the DATA are sufficiently reliable for the purposes of our work.

5. Conclusions

Our findings indicated that soils can take up CH₄ and N₂O from the atmosphere under dry and oxic conditions in low N input upland cropping systems. The 0–15 cm horizon contributed 98.2% of the cumulative surface CO₂ efflux, and the CH₄ and N₂O consumption occurred in the 0–40 and 0–15 cm soil horizons, respectively. The subsoil below 15 cm acted largely as a CO₂ buffer; the production/consumption potentials of CH₄ and N₂O were very weak below 40 and 15 cm, respectively. Our study further implied that the emissions of soil water-dissolved N₂O via plant transpiration may account for 13.0% of the surface exchange. Our findings also revealed that the GM is a suitable method to assess fluxes of CO₂ and the consumption of CH₄ in well aerated and horizontally homogeneous soils, while the CM is necessary to reduce the uncertainty arising from possible N₂O and CH₄ production in the top layer. Our findings improve the understanding of gas production or consumption processes in the soil-atmosphere continuum. Meanwhile, the GM coupled with chamber measurements can improve surface gas flux estimates for field and laboratory studies. Further studies are needed to explore the mechanisms responsible for changes in stored greenhouse gas within soil pore space and environmental factors driving these processes. This is of particular interest because these belowground gas pools may become increasingly important positive feedbacks as global climate disruption becomes more pronounced.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.geoderma.2018.01.001>.

Acknowledgements

This research was supported by the National Natural Science Foundation of China (Grant No. 41473021, Grant No. 41530859 and Grant No. 41571291) and the “Strategic Priority Research Program of Chinese Academy of Sciences” (Grant No. XDA05050601). It is also supported by Special Fund for Agro-scientific Research in the Public Interest (201503117-09). We thank X. H. Huang and X. F. Wang for hard work and much enthusiasm in the field, and O. Oenema for thoughtful discussions.

References

Banger, K., Tian, H.Q., Lu, C.Q., 2012. Do nitrogen fertilizers stimulate or inhibit methane emissions from rice fields? *Glob. Chang. Biol.* 18, 3259–3267.
 Bazylinski, D.A., Soohoo, C.K., Hollocher, T.C., 1986. Growth of *Pseudomonas aeruginosa* of nitrous-oxide. *Appl. Environ. Microbiol.* 51, 1239–1246.
 Bond-Lamberty, B., Thomson, A., 2010. A global database of soil respiration data. *Biogeosciences* 7, 1915–1926.

Bouyoucos, G.J., 1936. Directions for making mechanical analysis of soils by the hydrometer method. *Soil Sci.* 42 (3), 225–228.
 Bowden, W.B., Bormann, F.H., 1986. Transport and loss of nitrous oxide in soil water after forest cutting. *Science* 233, 867–869.
 Brasher, B.R., Franzmeier, D.P., Valassis, V.T., Davidson, S.E., 1966. Use of saran resin to coat natural soil clods for bulk density and water-retention measurements. *Soil Sci.* 101, 108.
 Burgos, M., Sierra, A., Ortega, T., Forja, J.M., 2015. Anthropogenic effects on greenhouse gas (CH₄ and N₂O) emissions in the Guadalete River Estuary (SW Spain). *Sci. Total Environ.* 503–504, 179–189.
 Castro, M.S., Melillo, J.M., Steudler, P.A., Chapman, J.W., 1994. Soil moisture as a predictor of methane uptake by temperate forest soils. *Can. J. For. Res.* 24 (9), 1805–1810.
 Chang, C., Janzen, H.H., Nakonechny, E.M., Cho, C.M., 1998. Nitrous oxide emission through plants. *Soil Sci. Soc. Am. J.* 62 (1), 35–38.
 Chapuis-Lardy, L., Wrage, N., Metay, A., Chotte, J.L., Bernoux, M., 2007. Soils, a sink for N₂O? A review. *Glob. Chang. Biol.* 13, 1–17.
 Chatskikh, D., Olesen, J.E., 2007. Soil tillage enhanced CO₂ and N₂O emissions from loamy sand soil under spring barley. *Soil Tillage Res.* 97 (1), 5–18.
 Dobbie, K.E., McTaggart, L.P., Smith, K.A., 1999. Nitrous oxide emissions from intensive agricultural systems: variations between crops and seasons, key driving variables, and mean emission factors. *J. Geophys. Res.-Atmos.* 104 (D21), 26891–26899.
 Dusenbury, M.P., Engel, R.E., Miller, P.R., Lemke, R.L., Wallander, R., 2008. Nitrous oxide emission from a northern Great Plains soil as influenced by nitrogen management and cropping systems. *J. Environ. Qual.* 37, 542–550.
 Fang, H.J., Cheng, S.L., Yu, G.R., Cooch, J., Wang, Y.S., Xu, M.J., Li, L.S., Dang, X.S., Li, Y.N., 2014. Low-level nitrogen deposition significantly inhibits methane uptake from an alpine meadow soil on the Qinghai–Tibetan Plateau. *Geoderma* 213, 444–452.
 Fest, B.J., Livesley, S.J., Dröslér, M., van Gorsel, E., Arndt, S.K., 2009. Soil–atmosphere greenhouse gas exchange in a cool, temperate *Eucalyptus delegatensis* forest in south-eastern Australia. *Agric. For. Meteorol.* 149, 393–406.
 Fierer, N., Chadwick, O.A., Trumbore, S.E., 2005. Production of CO₂ in soil profiles of a California annual grassland. *Ecosystems* 8, 412–429.
 Flechard, C.R., Neftel, A., Jocher, M., Ammann, C., Fuhrer, J., 2005. Bi-directional soil/atmosphere N₂O exchange over two mown grassland systems with contrasting management practices. *Glob. Chang. Biol.* 11, 2114–22127.
 Flessa, H., Ruser, R., Dörsch, P., Kamp, T., Jimenez, M.A., Munch, J.C., Beese, F., 2002. Integrated evaluation of greenhouse gas emissions (CO₂, CH₄, N₂O) from two farming systems in southern Germany. *Agric. Ecosyst. Environ.* 91, 175–189.
 Gebert, J., Röer, I.U., Scharff, H., Roncato, C.D.L., Cabral, A.R., 2011. Can soil gas profiles be used to assess microbial CH₄ oxidation in landfill covers? *Waste Manag.* 31, 987–994.
 Goldberg, S.D., Gebauer, G., 2009. Drought turns a Central European Norway spruce forest soil from an N₂O source to a transient N₂O sink. *Glob. Chang. Biol.* 15, 850–860.
 Guo, J.P., Zhou, C.D., 2007. Greenhouse gas emissions and mitigation measures in Chinese agroecosystems. *Agric. For. Meteorol.* 142, 270–277.
 Hou, L.Y., Wang, Z.P., Wang, J.M., Wang, B., Zhou, S.B., Li, L.H., 2012. Growing season in situ uptake of atmospheric methane by desert soils in a semiarid region of northern China. *Geoderma* 189–190, 415–422.
 Hutchinson, G.L., Livingston, G.P., 1993. Use of chamber systems to measure trace gas fluxes. In: Harper, L.A., Moiser, A.R., Duxbury, J.M., Rolston, D.E. (Eds.), *Agro-Ecosystem Effect on Radiatively Important Trace Gases and Climate Change*, vol. 55. American Society of Agronomy, Madison, Wis, USA, pp. 63–78.
 Hütsch, B.W., 2001. Methane oxidation in non-flooded soils as affected by crop production-invited paper. *Eur. J. Agron.* 14, 237–260.
 Kim, Y., Ueyama, M., Nakagawa, F., Tsunogai, U., Harazono, Y., Tanaka, N., 2007. Assessment of winter fluxes of CO₂ and CH₄ in boreal forest soils of central Alaska estimated by the profile method and the chamber method: a diagnosis of methane emission and implications for the regional carbon budget. *Tellus* 59B, 223–233.
 Lang, R., Blagodatsky, S., Xu, J.C., Cadisch, G., 2017. Seasonal differences in soil respiration and methane uptake in rubber plantation and rainforest. *Agric. Ecosyst. Environ.* 240, 314–328.
 Le Mer, J., Roger, P., 2001. Production, oxidation, emission and consumption of methane in soils: a review. *Eur. J. Soil Biol.* 37, 25–50.
 Lerman, A., 1979. *Geochemical Processes: Water and Sediment Environments*. Wiley, New York, pp. 481.
 Liebig, M.A., Tanaka, D.L., Gross, J.R., 2010. Fallow effects on soil carbon and greenhouse gas flux in central North Dakota. *Soil Sci. Soc. Am. J.* 74, 358–365.
 Livingston, G.P., Hutchinson, G.L., 1995. Enclosure-based measurement of trace gas exchange: application and sources of error. In: Matson, P.A., Harriss, R.C. (Eds.), *Biogenic Trace Gases: Measuring Emissions From Soil and Water*. Backwell Science, Cambridge, pp. 14–50.
 Lloyd, D., 1993. Aerobic denitrification in soils and sediments: from fallacies to facts. *Trends Ecol. Evol.* 8 (10), 352–356.
 Lohila, A., Aurela, M., Hatakka, J., Pihlatie, M., Minkinen, K., Penttilä, T., Laurila, T., 2010. Responses of N₂O fluxes to temperature, water table and N deposition in a northern boreal fen. *Eur. J. Soil Sci.* 61, 651–661.
 Maier, M., Schack-Kirchner, H., 2014. Using the gradient method to determine soil gas flux: a review. *Agric. For. Meteorol.* 192–193, 78–95.
 Maier, M., Schack-Kirchner, H., Hildebrand, E.E., Holst, J., 2010. Pore-space CO₂ dynamics in a deep, well-aerated soil. *Eur. J. Soil Sci.* 61, 877–887.
 Mast, M.A., Wickland, K.P., Striegl, R.T., Clow, D.W., 1998. Winter fluxes of CO₂ and CH₄ from subalpine soils in Rocky Mountain National Park, Colorado. *Glob. Biogeochem. Cycles* 12, 607–620.
 McLain, J.E.T., Martens, D.A., 2006. Moisture controls on trace gas fluxes in semiarid

- riparian soils. *Soil Sci. Soc. Am. J.* 70, 367–377.
- Millington, R., Quirk, J.P., 1961. Permeability of porous solids. *Trans. Faraday Soc.* 57, 1200–1207.
- Mosier, A., Schimel, D., Valentine, D., Bronson, K., Parton, W., 1991. Methane and nitrous oxide fluxes in native, fertilized and cultivated grasslands. *Nature* 350, 330–332.
- Mosier, A.R., Delgado, J.A., Keller, M., 1998. Methane and nitrous oxide fluxes in an acid oxisol in western Puerto Rico: effects of tillage, liming, and fertilization. *Soil Biol. Biochem.* 30, 2087–2098.
- Mu, Z., Huang, A., Kimura, S.D., Jin, T., Wei, S., Hatano, R., 2009. Linking N₂O emission to soil mineral N as estimated by CO₂ emission and soil C/N ratio. *Soil Biol. Biochem.* 41, 2593–2597.
- Nakano, T., Inoue, G., Fukuda, M., 2004. Methane consumption and soil respiration by a birch forest soil in West Siberia by a birch forest soil in West Siberia. *Tellus* 56B, 223–229.
- Nan, W.N., Yue, S.C., Li, S.Q., Huang, H.Z., Shen, Y.F., 2016. Characteristics of N₂O production and transport within soil profiles subjected to different nitrogen application rates in China. *Sci. Total Environ.* 542, 864–875.
- Ojima, D.S., Valentine, D.W., Mosier, A.R., Parton, W.J., Schimel, D.S., 1993. Effect of land use change on methane oxidation in temperate forest and grassland soils. *Chemosphere* 26 (1–4), 675–685.
- Patureau, D., Bernet, N., Delgenes, J.P., Moletta, R., 2000. Effect of dissolved oxygen and carbon-nitrogen loads on denitrification by an aerobic consortium. *Appl. Microbiol. Biotechnol.* 54, 535–542.
- Petersen, S.O., Mutegei, J.K., Hansen, E.M., Munkholm, L.J., 2011. Tillage effects on N₂O emissions as influenced by a winter cover crop. *Soil Biol. Biochem.* 43, 1509–1517.
- Pihlatie, M., Ambus, P., Rinne, J., Pilegaard, K., Vesala, T., 2005. Plant-mediated nitrous oxide emissions from beech (*Fagus sylvatica*) leaves. *New Phytol.* 168, 93–98.
- Pritchard, D.T., Currie, J.A., 1982. Diffusion of coefficients of carbon-dioxide, nitrous-oxide, ethylene and ethane in air and their measurement. *J. Soil Sci.* 33, 175–184.
- Reichstein, M., Beer, C., 2008. Soil respiration across scales: the importance of a model-data integration framework for data interpretation. *J. Plant Nutr. Soil Sci.* 171, 344–354.
- Reth, S., Graf, W., Gefke, O., Schilling, R., Seidlitz, H.K., Munch, J.C., 2008. Whole year-round observation of N₂O profiles in soil: a lysimeter study. *Water Air Soil Pollut. Focus* 8, 129–137.
- Rochette, P., Hutchinson, G.L., 2004. Measurement of soil respiration in situ: chamber techniques. In: Hatfield, J.L. (Ed.), *Micrometeorology in Agriculture*. ASA, CSSA and SSSA, Madison, WI.
- Roobroek, D., Butterbach-Bahl, K., Brüggemann, N., Boeckx, P., 2010. Dinitrogen and nitrous oxide exchanges from an undrained monolith fen: short-term responses following nitrate addition. *Eur. J. Soil Sci.* 61, 662–670.
- Ruan, L., Robertson, G.P., 2013. Initial nitrous oxide, carbon dioxide, and methane costs of converting Conservation Reserve Program grassland to row crops under no-till vs. conventional tillage. *Glob. Chang. Biol.* 19, 2478–2489.
- Sallam, A., Jury, W.A., Letey, J., 1984. Measurement of gas-diffusion coefficient under relatively low air-filled porosity. *Soil Sci. Soc. Am. J.* 48, 3–6.
- Sanz-Cobena, A., Sánchez-Martín, L., García-Torres, L., Vallejo, A., 2012. Gaseous emissions of N₂O and NO and NO₃⁻ leaching from urea applied with urease and nitrification inhibitors to a maize (*Zea mays*) crop. *Agric. Ecosyst. Environ.* 149, 64–73.
- Sanz-Cobena, A., García-Marco, S., Quemada, M., Gabriel, J.L., Almendros, P., Vallejo, A., 2014. Do cover crops enhance N₂O, CO₂ or CH₄ emissions from soil in Mediterranean arable systems? *Sci. Total Environ.* 466–467, 164–174.
- Soil Survey Staff, 2006. *Keys to Soil Taxonomy*, tenth ed. USDA-Natural Resources Conservation Service, Washington, D.C.
- Sparks, D.L., 1996. *Methods of soil analysis, part 3—chemical methods*. In: Sparks, D.L. (Eds.), *SSSA Book Series, No. 5*. SSSA, Inc. and American Society of Agronomy, Inc., Madison, WI, pp. 475–1185.
- Stuedler, P.A., Bowden, R.D., Melillo, J.M., Aber, J.D., 1989. Influence of nitrogen fertilization on methane uptake in temperate forest soils. *Nature* 341, 314–316.
- Stewart, K.J., Brummell, M.E., Farrell, R.E., Siciliano, S.D., 2012. N₂O flux from plant-soil systems in polar deserts switch between sources and sinks under different light conditions. *Soil Biol. Biochem.* 48, 69–77.
- Tang, J.W., Baldocchi, D.D., Qi, Y., Xu, L.K., 2003. Assessing soil CO₂ efflux using continuous measurements of CO₂ profiles in soils with small solid-state sensors. *Agric. For. Meteorol.* 118, 207–220.
- Valentini, R., Matteucci, G., Dolman, A.J., Schulze, E.D., Rebmann, C., et al., 2000. Respiration as the main determinant of carbon balance in European forests. *Nature* 404, 861–865.
- Wang, Y.Y., Hu, C.S., Ming, H., Zhang, Y.M., Li, X.X., Dong, W.X., Oenema, O., 2013. Concentration profiles of CH₄, CO₂ and N₂O in soils of a wheat–maize rotation ecosystem in North China Plain, measured weekly over a whole year. *Agric. Ecosyst. Environ.* 164, 260–272.
- Wang, Y.Y., Hu, C.S., Ming, H., Oenema, O., Schaefer, D.A., Dong, W.X., Zhang, Y.M., Li, X.X., 2014. Methane, carbon dioxide and nitrous oxide fluxes in soil profile under a winter wheat–summer maize rotation in the North China Plain. *PLoS One* 9 (6), e98445.
- Whalen, S.C., Reeburgh, W.S., 1990. Consumption of atmospheric methane by tundra soils. *Nature* 346, 160–162.
- Wu, D.M., Dong, W.X., Oenema, O., Wang, Y.Y., Trebs, I., Hu, C.S., 2013. N₂O consumption by low-nitrogen soil and its regulation by water and oxygen. *Soil Biol. Biochem.* 60, 165–172.
- Ye, R.Z., Horwath, W., 2016. Nitrous oxide uptake in rewetted wetlands with contrasting soil organic carbon contents. *Soil Biol. Biochem.* 100, 110–117.
- Zausig, J., Stepniowski, W., Horn, R., 1993. Oxygen concentration and redox potential gradients in unsaturated model soil aggregates. *Soil Sci. Soc. Am. J.* 57, 908–916.
- Zhao, N., Meng, P., Zhang, J., Lu, S., Cheng, Z., 2014. Comparison of soil respiration under various land uses in hilly area of Northern China. *Sci. Silvae Sin.* 50, 1–7 (in Chinese).