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Concentration profiles of CH_4, CO_2 and N_2O in soils of a wheat–maize rotation ecosystem in North China Plain, measured weekly over a whole year

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ABSTRACT

Agricultural soils are main sources and sinks of the greenhouse gases carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). The source–sink function depends on soil characteristics, climate and management. Emission measurements usually quantify the net result of production, consumption and transport of these gases in the soil; they do not provide information about the depth distributions of the concentrations of these gases in the soil. Here we report on concentrations of CO_2, CH_4 and N_2O in air of 300 cm deep soil profiles, at resolutions of 30–50 cm, over a full year. Gas samples were taken weekly in a long-term field experiment with an irrigated winter wheat–summer maize double cropping system, and four fertilizer N application rates (0, 200, 400 and 600 kg N ha^{-1} year^{-1}). The results showed distinct differences in CH_4, CO_2 and N_2O concentrations profiles with soil depth. The concentrations of CO_2 in soil air increased with soil depth and showed a seasonal pattern with relatively high concentrations in the warm and moist maize growing season and relatively low concentrations in the winter-wheat growing season. In contrast, CH_4 concentrations decreased with depth, and did not show a distinct seasonal cycle. Urea application did not have a large effect on CH_4 or CO_2 concentrations, neither in the topsoil nor the subsoil. Concentrations of N_2O responded to N fertilizer application and irrigation. Application of fertilizer strongly increased grain and straw yields of both winter wheat and summer maize, relatively to the control, but differences in yield between the treatments N200, N400 and N600 were not statistically significant. However, it significantly increased mean N_2O concentrations peaks at basically all soil depths. Interestingly, concentrations of N_2O increased almost instantaneously in the whole soil profile, which indicates that the soil had a relatively high diffusivity, despite compacted subsoil layers.

In conclusion, the frequent measurements, at high depth resolutions, of concentrations of CH_4, CO_2 and N_2O in soil air under a winter wheat–summer maize double crop rotation provide detailed insight into the production, consumption and transport of these gases in the soil. Concentrations of CH_4, CO_2 and N_2O responded differently to management activities and weather conditions.

1. Introduction

During the past few decades, atmospheric concentrations of methane (CH_4), carbon dioxide (CO_2) and nitrous oxide (N_2O) have been increasing at rates of 0.8, 0.5 and 0.3% year^{-1}, respectively. The rising concentrations of these greenhouse gases (GHG) have been implicated with global climate change (IPCC, 2007). Worldwide concerns about climate change and its effects on our future environment has provided a strong impetus to research aimed at a better understanding of the cause–effect relationships of GHG emissions (Mosier et al., 1998; Dalal and Allen, 2008). Agriculture plays an important role in the global flux of GHG gases. The increasing demand for food by the increasing human population has incentivized the intensification of agricultural production. Land use changes, irrigation, drainage and fertilizer application practices and changes in crop and livestock patterns have contributed to changes in carbon (C) and nitrogen (N) dynamics and thereby also to changes in GHG emissions. Especially soils are major sources of GHG emissions.

It has been well documented that applications of fertilizers and animal manures not only increase the emissions of N_2O but also CO_2, and CH_4 from soils (Hütsch et al., 1994; Li et al., 1997; Drury et al., 1998; Matson et al., 1998; Ruser et al., 2001, 2006; Scheer et al., 2008). The GHG emissions from soils are the result of complex production, consumption and transport processes, and are affected by a range of environmental and management factors. Measurements of the GHG emissions at the soil surface provide an integrative estimate

Table 1
Soil characteristics at the experimental site in 2007.

Depth (cm)	pH (H_2O)	Sand (%)	Silt (%)	Clay (%)	Textural class[a]	Dry bulk density ($g\,cm^{-3}$)	Total organic matter ($g\,kg^{-1}$)	Total nitrogen ($g\,N\,kg^{-1}$)	Available nitrogen (mg $N\,kg^{-1}$)	Available phosphorus (mg $P_2O_5\,kg^{-1}$)	Available potassium (mg $K_2O\,kg^{-1}$)
0–30	8.5	25	58	17	SSL	1.47	16.7	1.40	148	9.3	95.6
30–60	7.7	22	60	18	L	1.40	10.9	0.85	72.0	4.9	65.6
60–90	7.8	31	55	14	L	1.45	7.8	0.64	70.1	1.0	44.5
90–150	7.8	15	59	26	SCL	1.57	6.5	0.38	49.8	0.4	33.5
150–200	7.7	18	47	35	GCL	1.43	5.4	0.28	38.9	0.3	25.6
200–250	7.8	15	35	50	C	1.51	4.2	0.15	30.1	0.2	16.8
250–300	7.8	12	35	53	C	1.50	3.0	0.86	25.9	0.1	8.5

[a] SSL, silty sandy loam; L, loam; SCL, silty clay loam; GCL, gravely clay loam; C, clay.

of the net result of the production, consumption and transport in soil but provide no information about the CH_4, CO_2 and N_2O production–consumption–transport patterns within soil profiles. Understanding these patterns might provide a better insight into the possibilities to reduce GHG emissions. For example, a temporary accumulation of GHG in the soil profile influences GHG flux patterns at the soil surface over time, and thereby may confuse empirical relationships between agricultural activities and measured GHG emissions (Bowden and Bormann, 1986). Thus, measurements of CH_4, CO_2 and N_2O concentration gradients in the soil profile may be of benefit for estimating and understanding their fluxes at the soil surface.

The production and consumption of GHG in soils are related to microbiological processes and the roles of microorganisms and their controlling factors are very important therefore (Chen et al., 2010). Their activities are controlled by environmental conditions, including temperature, rainfall and soil biological, chemical and physical characteristics. As a result, emissions of GHG from soils have been related to climate, management activities (e.g. soil cultivation, irrigation, fertilizer application), and various soil characteristics, e.g. soil organic carbon and nitrogen contents, dissolved organic C and N contents, mineral N contents, soil bulk density, salinity and redox potential (Huang et al., 2009). However, the relationships between GHG emissions and climate, managements and soil characteristics are often confused by huge spatial and temporal variations in emissions, in part because of complex interactions between GHG productions–consumptions–transports in the soil profile (Wang et al., 2005; Panikov et al., 2007).

The general hypothesis of the study reported here is that measurements of N_2O, CH_4 and CO_2 concentration gradients in soil profiles may contribute to a better understanding of the temporal variation in the soil–atmosphere exchanges of these gases. So far, only few investigations have determined N_2O, CH_4 and CO_2 concentration gradients in soil profiles, and very few have reported measurements all three major GHG gases simultaneously (Van Groenigen et al., 2005; Reth et al., 2008; Liang et al., 2009). Hence, the objectives of the study reported here were (1) to determine the seasonal variations in the concentrations of both N_2O, CH_4 and CO_2 in soil from the soil surface to a depth of 300 cm, and (2) to analyze the effects of agricultural activities (N fertilizer application, irrigation) and soil moisture and temperature on the concentrations of N_2O, CH_4 and CO_2 in soil. Measurements were carried out in a winter wheat-summer maize double cropping system in the North China Plain, which is a main agro-ecosystem.

2. Materials and methods

2.1. Site characteristics

The study was conducted at the Luancheng Agroecosystem Experimental Station (37°53′N, 114°41′E, elevation 50 m) of the Chinese Academy of Sciences. The station is at the piedmont of the Taihang Mountains, in the North China Plain. Mean annual precipitation is about 480 mm, 70% of which is in the period July to September. Annual average air temperature is 12.5 °C. The soil type of the area is predominantly silt loam (light meadow cinnamon soil). Soil characteristics of the experimental field are listed in Table 1.

The dominant cropping system in the region is a winter wheat-summer maize double cropping system (two crop harvests in a single year) without fallow between the crops. Commonly, winter wheat is sown in October and harvested in June; summer maize is sown almost immediately after the wheat harvest and harvested in October. Both wheat and maize are irrigated, mainly via flood irrigation using groundwater.

2.2. Field experimental design

Measurements were carried out during the 2007–2008 growing seasons, in an existing long-term winter wheat-summer maize double-cropping field experiment. The field experiment started in 1998 and had four treatments in triplicate in a randomized complete block design. Treatments included four N fertilizer application rates: 0 (control, N0), 200 (N200), 400 (N400) and 600 (N600) kg N ha^{-1} $year^{-1}$. Plot size was 7 m × 10 m.

Details on fertilizer application and crop husbandry are presented in Tables 2 and 3, respectively. Crops are flood-irrigated with pumped groundwater. At harvest, grain and straw yields were determined in three 1 m^2 areas in each plot. Straw was chopped (<5 cm) by automated machine and returned to the field.

Daily rainfall and mean daily air temperature were recorded at a weather station on the experimental site. Soil temperature was measured using seven CS107b soil temperature probes (Cambell Scientific Inc., Logan, UT) installed at soil depth of 30, 60, 90, 150, 200, 250 and 300 cm. Three-meter neutron access tubes were installed at each plot. Soil moisture was measured using a neutron moisture meter.

2.3. Soil gas sampling and measurements

In 2006, 8 years after the start of the experiment, seven soil–air equilibration samplers (Fig. 1) were installed in each plot at soil depth of 25–30, 55–60, 85–90, 145–150, 195–200, 245–250 and 295–300 cm. (For convenience, we use the bottom depth below each layer representing the whole observed layer in the following figures and tables.) These samplers were constructed from polyvinylchloride (PVC) tubes with a 2.5 cm inner diameter. The lower part of the PVC tube was perforated and covered by a water impermeable membrane, which allows 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. The soil–air equilibration tubes were carefully inserted in pre-drilled holes, made by a 3.0 cm diameter hand auger. The spaces around the tubes were backfilled with soil in the

Table 2
Fertilization treatments.

Treatments	Basal fertilization, applied at wheat sowing (kg ha^{-1})			Supplementary N fertilization (kg ha^{-1})	
	N	P$_2$O$_5$	K$_2$O	Wheat (in March)	Maize (in July)
N0	0	65	0	0	0
N200	50	65	0	50	100
N400	100	65	0	100	200
N600	150	65	0	150	300

Table 3
Timing of crop management activities.

Timing	Crop management activities	
	Winter-wheat season	Summer-maize season
October 3, 2006	Basal N fertilization and irrigation	
October 10, 2006	Seeding	
April 7, 2007	Supplementary N fertilization and irrigation	
May 19, 2007	Irrigation	
June 1, 2007		Seeding
June 14, 2007	Harvest	
June 19, 2007		Irrigation
July 27, 2007		Supplementary N fertilization
July 29, 2007		Irrigation
October 1, 2007		Harvest
October 2, 2007	Basal N fertilization and irrigation	
October 7, 2007	Seeding	

same order as it was dug out. A tiny rod was used to gently press the soil around the tubes.

Soil–air sampling started 4 months after the installation of the samplers. Samples were taken twice a week between 9:00 AM and 11:00 AM, using 100 ml plastic syringes connected to the tubes via the three-way stopcocks at the surface. The surface air was concurrently sampled. The gas samples were analyzed by a gas chromatography (Agilent GC-6820, Agilent Technologies Inc., Santa Clara, CA, USA) with separate electron capture and flame ionization detectors (ECD at 330 °C and FID at 200 °C) for N_2O, CH_4 and CO_2, respectively.

2.4. Soil analyses

Soil particle size analysis was done by the Bouyoucos Hydrometer Method (Bouyoucos, 1936). Soil bulk density was determined using the cutting ring method (Brasher et al., 1966). Soil pH was measured in a suspension of 5 g soil with 25 ml distilled water, 1 h after shaking. 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.

2.5. Data analyses

All data were subjected to statistical analysis (SPSS 13.0). Data were analyzed using ANOVA. Differences between treatment means were compared using Fisher's LSD method at the 0.05 probability level.

3. Results

3.1. Soil water content and soil temperature

Changes in soil water content with depth are shown in Fig. 2. Irrigation and rainfall events increased soil water content at all depths. There were no statistical significant differences between fertilizer application treatments in soil water content, neither in the topsoil nor the subsoil. Following the irrigation events (see Table 3) on April 7 (94 mm), May 19 (60 mm), June 19 (60 mm) and July 29 (60 mm), water filled pore space (WFPS) in the soil increased significantly. Rainfall events on 30 April (17 mm), from 21 to 22 May (42 mm) and between 29 June and 21 August (204 mm) also increased soil moisture contents at various depths (Fig. 2).

Soil temperature variations showed clear seasonal patterns, with highest temperature during the maize growing season (Fig. 3, Table 3). The amplitude of the seasonal variations was about 20 °C in the top 30 cm and 5–10 °C at depth of 200–300 cm. At 30 cm, lowest temperature was 2 °C.
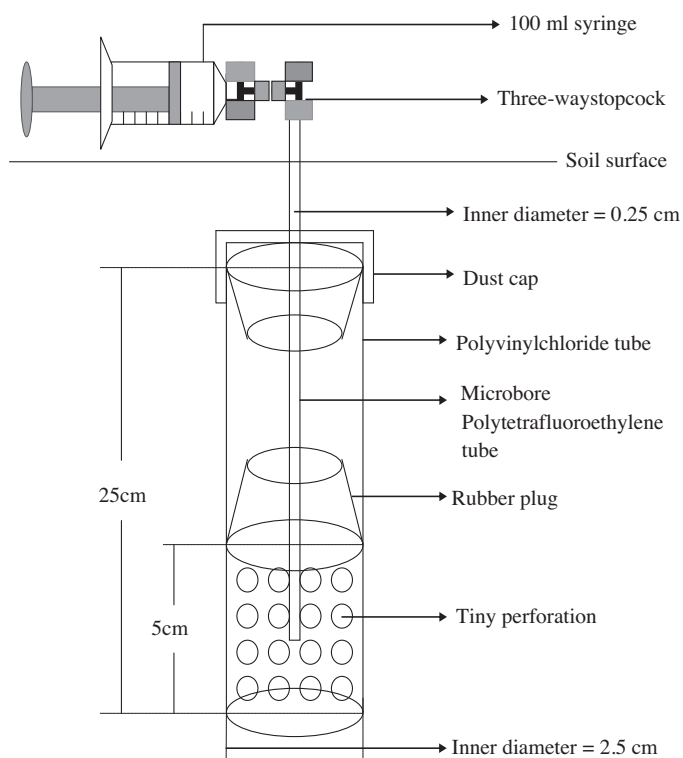
Fig. 1. The soil–air equilibration tube and the syringe (on top), when connected during sampling.
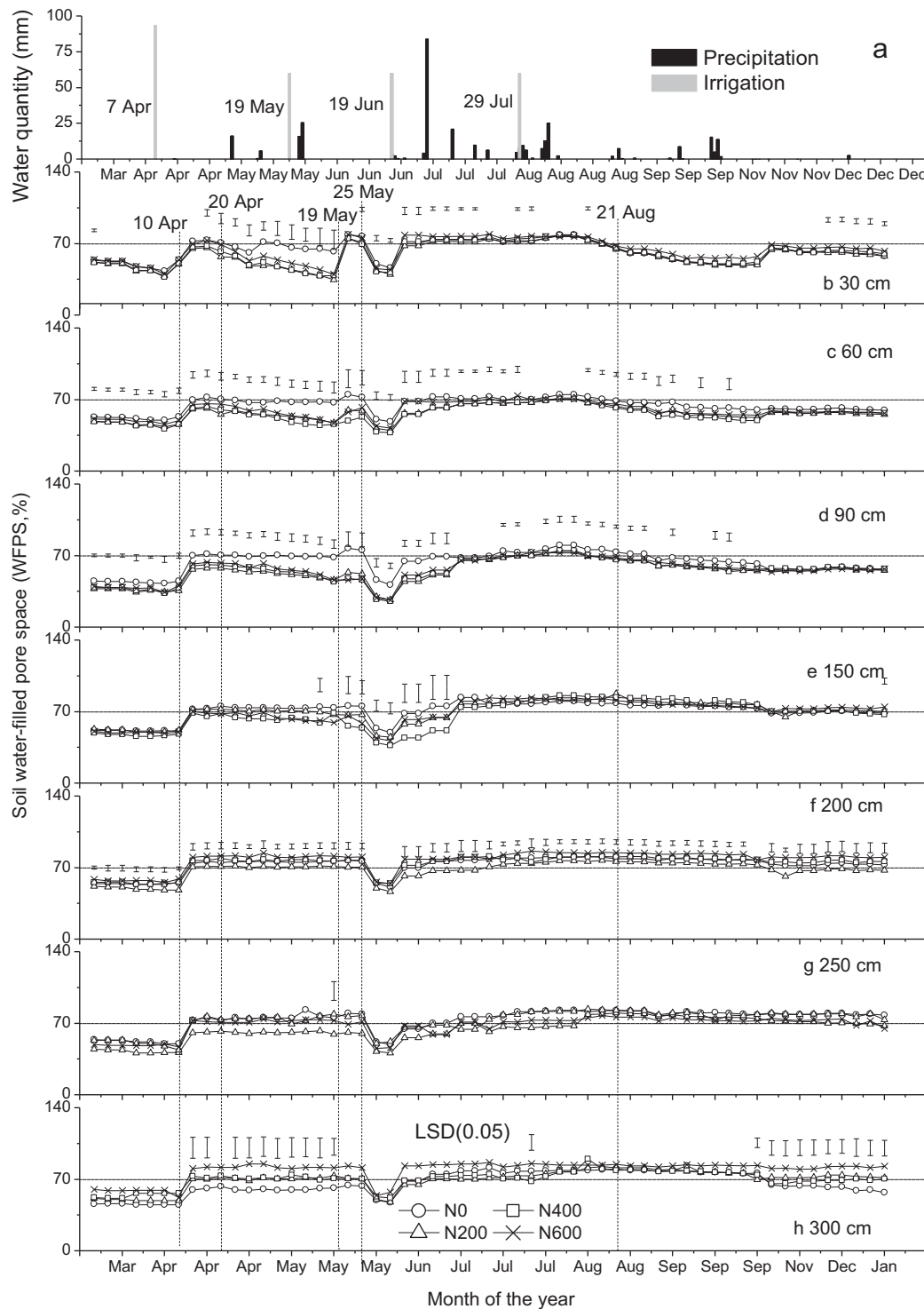
Fig. 2. Precipitation and irrigation events (top panel) and water-filled pore space (WFPS) at various soil depths (lower panels) in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha^{-1} year^{-1}, in 2007–2008. Bars in figures indicate 1 standard deviation ($n = 3$).

3.2. Maize and wheat yields

Mean maize and wheat grain and straw yields are presented in Table 4. Application of 100 kg N per hectare to both wheat and maize (N200 treatment) tripled grain yields and doubled straw yields of both wheat and maize, relative to the unfertilized control. On average, yields were highest in the N600 treatment, but differences between the N200, N400 and N600 treatments in grain and straw yield were not always statistical significant (Table 4).

Fertilizer N application increased total annual grain yield from 4500 kg per hectare in the control to almost 15,000 kg per hectare.

3.3. Concentration profiles of CH$_4$ in soil

Mean concentrations of CH$_4$ in soil air steadily decreased with depth (Fig. 4, Table 5A). Measured ambient air concentrations at the site ranged from 1.85 to 2.99 ml m^{-3}; at a depth of 30 cm, CH$_4$ concentrations ranged between 0.47 and 2.27 ml m^{-3}, suggesting
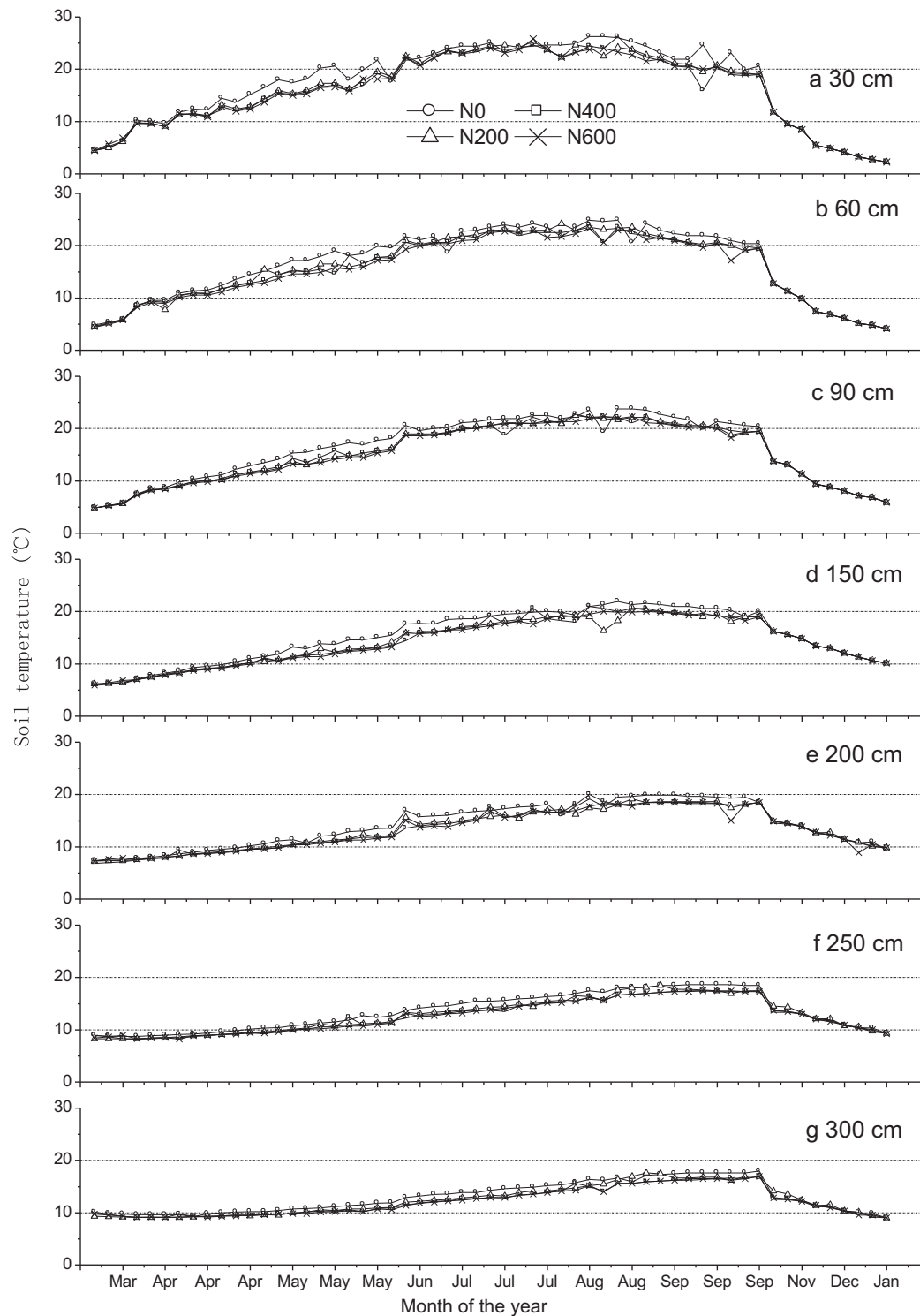
Fig. 3. Soil temperatures at various soil depths in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha^{-1} year^{-1}, in 2007–2008.

Table 4
Grain and straw yield of wheat and maize in 2007, in kg ha^{-1}.

Treatments	Wheat yield				Maize yield			
	Straw	C.V.	Grain	C.V.	Straw	C.V.	Grain	C.V.
N0	2803c	3	2357c	6	2194b	4	2353b	2
N200	7904b	7	7410ab	4	4732a	5	6409a	9
N400	7722b	6	7151b	9	4807a	3	6473a	1
N600	9109a	3	7855a	1	5613a	11	7211a	8

Values within the same column followed by the same letter are not significantly different as determined by ANOVA and Fisher protected LSD ($P < 0.05$).
C.V., coefficient of variance (%).

Table 5
Average concentration of CH_4 (A), CO_2 (in ml m^{-3}) (B), and N_2O (in μl m^{-3}) (C).

(A) CH_4

Treatments	30 cm	60 cm	90 cm	150 cm	200 cm	250 cm	300 cm
N0	1.56 (3)a	0.68 (4)a	0.58 (12)a	0.40 (4)a	0.36 (14)a	0.43 (2)a	0.37 (7)a
N200	1.53 (6)a	0.50 (16)b	0.30 (5)b	0.31 (13)b	0.28 (6)a	0.33 (1)a	0.27 (8)ab
N400	1.47 (8)a	0.51 (2)b	0.29 (6)b	0.28 (7)b	0.37 (3)a	0.48 (7)a	0.28 (11)ab
N600	1.45 (5)a	0.40 (6)b	0.31 (12)b	0.29 (14)b	0.28 (6)a	0.28 (4)a	0.26 (11)b

(B) CO_2

Treatments	30 cm		60 cm		90 cm		150 cm		200 cm		250 cm		300 cm	
	Wheat	Maize	Wheat	Maize	Wheat	Maize	Wheat	Maize	Wheat	Maize	Wheat	Maize	Wheat	Maize
N0	7038 (3)ab	10,160 (5)a	11,060 (12)a	23,425 (8)a	11,934 (7)b	24,427 (10)a	13,012 (2)b	26,445 (3)a	13,799 (3)ab	26,656 (3)a	14,190 (2)b	26,218 (1)a	15,846 (13)a	27,561 (2)a
N200	5338 (4)b	12,131 (8)a	11,640 (5)a	22,600 (10)a	14,076 (6)a	24,738 (5)a	15,037 (5)a	24,790 (2)a	14,873 (6)a	25,430 (6)ab	16,282 (9)a	26,725 (6)a	17,277 (4)a	25,917 (6)a
N400	7738 (11)a	9165 (2)a	11,606 (5)a	22,184 (6)a	13,790 (5)a	24,167 (6)a	14,382 (3)a	25,057 (6)a	13,348 (6)b	23,276 (6)b	13,170 (8)b	20,963 (5)a	15,226 (4)a	22,849 (1)b
N600	7866 (5)a	12,138 (5)a	12,940 (13)a	22,237 (5)a	12,780 (7)ab	23,904 (4)a	12,868 (9)b	24,204 (6)a	12,977 (6)b	24,148 (6)b	13,717 (8)b	22,239 (4)a	15,116 (4)a	21,993 (10)b

(C) N_2O

Treatments	30 cm		60 cm		90 cm		150 cm		200 cm		250 cm		300 cm	
	Peak	No peak	Peak	No peak	Peak	No peak	Peak	No peak	Peak	No peak	Peak	No peak	Peak	No peak
N0	1321 (6)b	462 (7)b	2480 (8)c	611 (13)b	2612 (5)c	614 (12)c	2659 (15)d	677 (11)c	2890 (11)d	731 (13)c	2867 (10)b	809 (1)b	3627 (14)c	1070 (2)b
N200	3733 (4)ab	580 (4)b	3883 (4)bc	801 (14)b	4154 (4)c	822 (9)bc	4334 (6)c	874 (4)bc	4395 (4)c	923 (3)bc	4806 (7)ab	1082 (9)b	4410 (5)bc	1108 (7)b
N400	3307 (6)ab	762 (4)ab	5915 (3)ab	980 (1)b	6074 (14)b	993 (13)b	6066 (8)b	1064 (11)b	6410 (11)b	1073 (5)b	5237 (3)ab	1129 (6)b	5769 (3)ab	1371 (11)b
N600	10,538 (7)a	1132 (11)a	7599 (10)a	1455 (3)a	8231 (10)a	1421 (4)a	7913 (9)a	1520 (3)a	7881 (8)a	1579 (2)a	7059 (2)a	1767 (1)a	6709 (7)a	1983 (3)a

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 ($P < 0.05$).
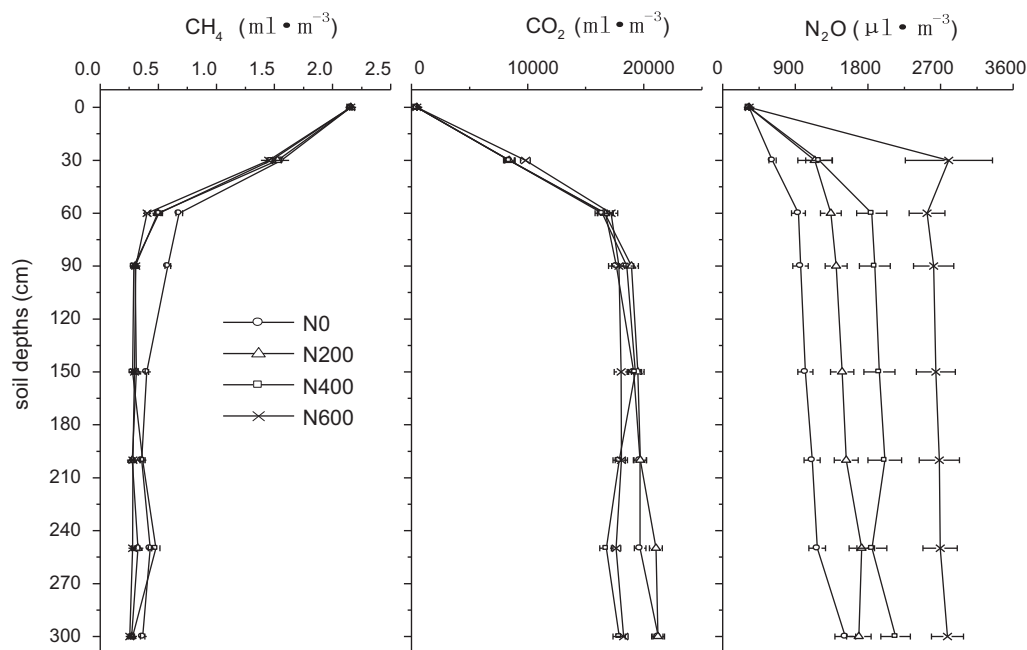
Fig. 4. Methane, carbon dioxide and nitrous oxide concentrations (mean ± standard deviations, $n = 57$) in soil air at various soil depths in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha^{-1} year^{-1}, in 2007–2008.

consumption of CH_4 by methanotrophic bacteria in the topsoil. Concentrations of CH_4 tended to decrease further between 30 and 60 cm, but remained more or less constant between 60 and 300 cm, in the range of 0.02–1.31 ml m^{-3} (Fig. 4, Table 5A). During the winter wheat growing season (Table 3), concentrations of CH_4 tended to decrease also between depths of 60 and 150 cm (Fig. 5). However, it cannot be excluded that this trend is related to equilibration processes following the installation of the gas samplers in the soil.

There were no clear seasonal patterns in the variations of CH_4 concentrations profiles (Fig. 5). Variations in CH_4 concentrations tended to be smaller during the wet maize growing season than during the dry and cold wheat growing season (Table 3, Fig. 5). Short-term temporal increases in CH_4 concentrations in soil, for example, at the transition of the wheat and maize growing seasons may be related to enhanced diffusion of CH_4 into the dry soil.

Urea application decreased annual mean CH_4 concentrations statistically significant (Table 5A) in both the topsoil and the subsoil. Following the irrigation and supplementary fertilizer N application on 7 April, there were no major changes in the concentrations of CH_4 in soil at 30–300 cm. Also later in the season, there were no large changes in the concentrations of CH_4 in soil air following rainfall, irrigation and fertilizer application events, apart perhaps from the heavy rainfall event in June and July (Fig. 5).

3.4. Concentration profiles of CO_2 in soil

Concentrations of CO_2 increased with soil depth (Fig. 4, Table 5B). Measured ambient air concentrations at the site ranged from 388 to 631 ml m^{-3}; at a depth of 30 cm, CO_2 concentration ranged between 1300 and 23,000 ml m^{-3} and at 60 cm between 4000 and 36,000 ml m^{-3} suggesting a significant respiration by soil bacteria in the upper 60 cm of soil (Figs. 4 and 6). Below 60 cm, CO_2 concentrations remained more or less constant (Fig. 4).

Urea application did not have statistically significant effects on annual mean CO_2 concentrations in either topsoil or subsoil (Table 5B). However, there were clear seasonal variations in CO_2 concentrations profiles. Concentrations of CO_2 were higher during the wet and warm maize growing season than during the dry and cold wheat growing season (Tables 3 and 5B, Fig. 6). With

increasing soil depth, concentration changes occurred somewhat later in the season. Incidental concentration changes were related to heavy rainfall and irrigation events. For example, concentration of CO_2 increased at 30 cm from 10 to 20 April and from 30 to 300 cm between 31 July and 17 August following irrigation and N fertilizer application on 7 April and from 27 to 29 July, respectively (see Fig. 6). However, later irrigation and heavy rainfall events induced temporary drops in CO_2 concentrations, at all depths from 17 to 28 August (see Fig. 6). Apparently, the diffusive efflux from the topsoil to the atmosphere was strong enough to limit the build-up of high CO_2 concentrations in the topsoil. In contrast, the diffusive efflux of CO_2 from the subsoil to the atmosphere is constrained by a relatively high bulk density in the subsoil (Table 1). At greater depth, changes in CO_2 concentrations are damped; concentrations remain relatively high during the wheat growing season while the peaks during the maize growing season are somewhat less and occur later. Apparently, nitrogen application and irrigation did not greatly affect root growth and respiration in the subsoil.

3.5. Concentration profiles of N_2O in soil

Concentration profiles of N_2O in soil (Figs. 4 and 7) were considerably influenced by the main management activities outlined in Table 3, such as irrigation and nitrogen applications. The supplementary fertilizer N applications in April and July induced peaks of N_2O concentrations throughout the soil profile. Likely, peaks were enhanced by the subsequent irrigation events. Maximum peaks were recorded at a depth of 30 cm. Peak height increased with N fertilizer application rate (Table 5C, Fig. 7).

After the supplementary fertilizer N application and irrigation on 7 April, the N_2O concentration peaked on 10 April at depth of 30–60 cm and on 14 April at depth of 90–300 cm (see Fig. 7). After the irrigations on 19 May (60 mm) and 19 June (60 mm) (Fig. 2), N_2O concentration peaks were observed on 23 May at depth of 30–60 cm and 22 June at depth of 30 cm, respectively (see Fig. 7). The supplementary fertilizer N applications and irrigation at the end of July induced peaks of N_2O from the end of July till
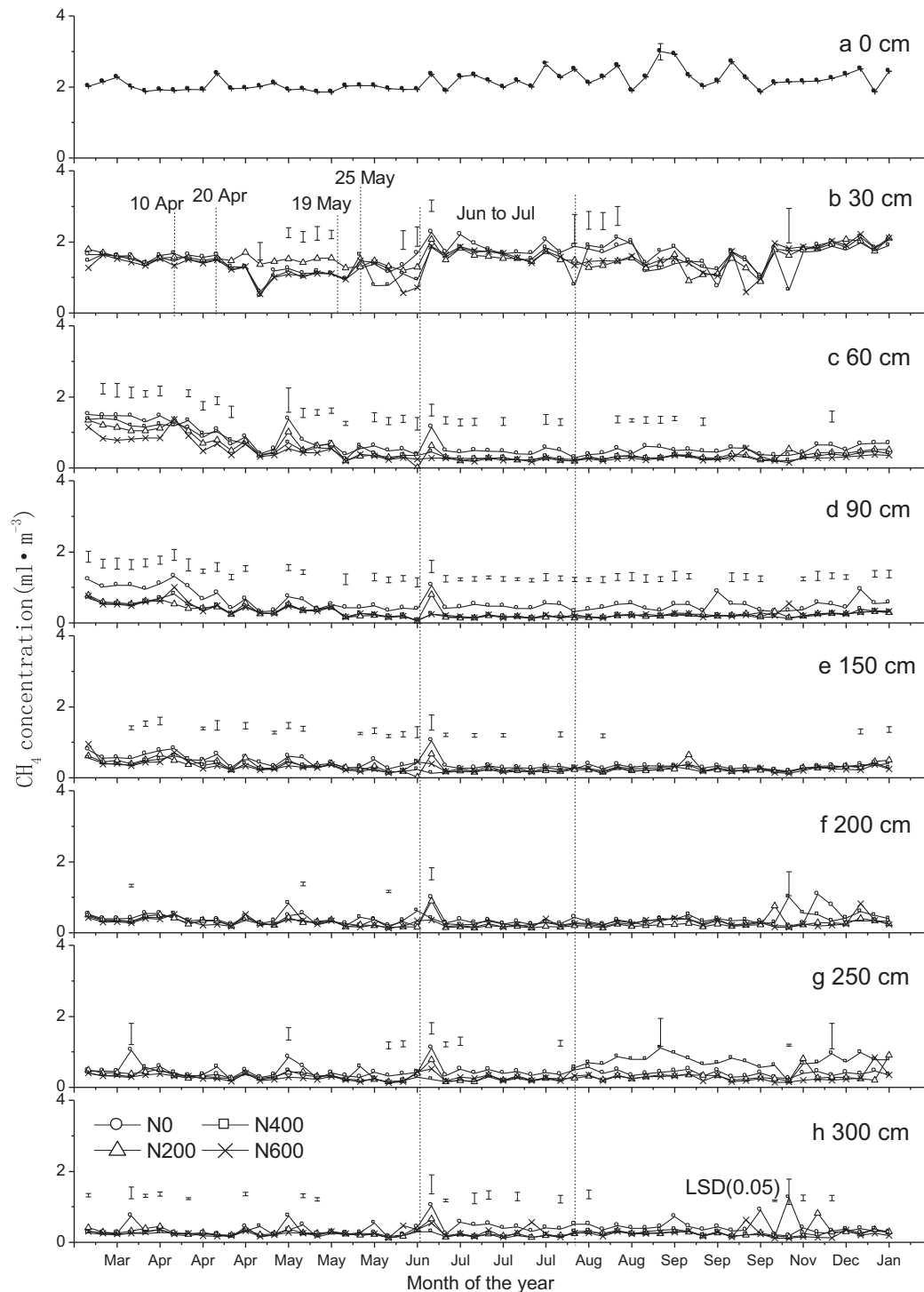
Fig. 5. Methane concentrations in soil air (in ml per m^3) at various soil depths in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha^{-1} year^{-1}, in 2007–2008. Bars in figures indicate 1 standard deviation ($n = 3$).

mid-August. Simultaneously, there was a delay effect in the occurrence of peak concentration with soil depth (see Fig. 7).

4. Discussion

4.1. Main findings

This is one of the first studies presenting weekly measurements of CH$_4$, CO$_2$ and N$_2$O concentrations in soil air down to a depth of 300 cm, at intervals of 30 to 50 cm, and for more than

a year. We found distinct differences in CH$_4$, CO$_2$ and N$_2$O concentrations profiles with soil depth in the winter wheat–summer maize double cropping system. Concentrations of CO$_2$ in soil followed a distinct seasonal cycle with relatively high concentrations during the warm and wet maize growing season and relatively low concentrations during the cold and dry winter growing season. In contrast, no clear seasonal patterns were found for the concentration profiles of CH$_4$ and N$_2$O. However, management activities and heavy rainfall events induced incidental changes in the concentrations of CH$_4$, CO$_2$ and N$_2$O at basically all soil depths.
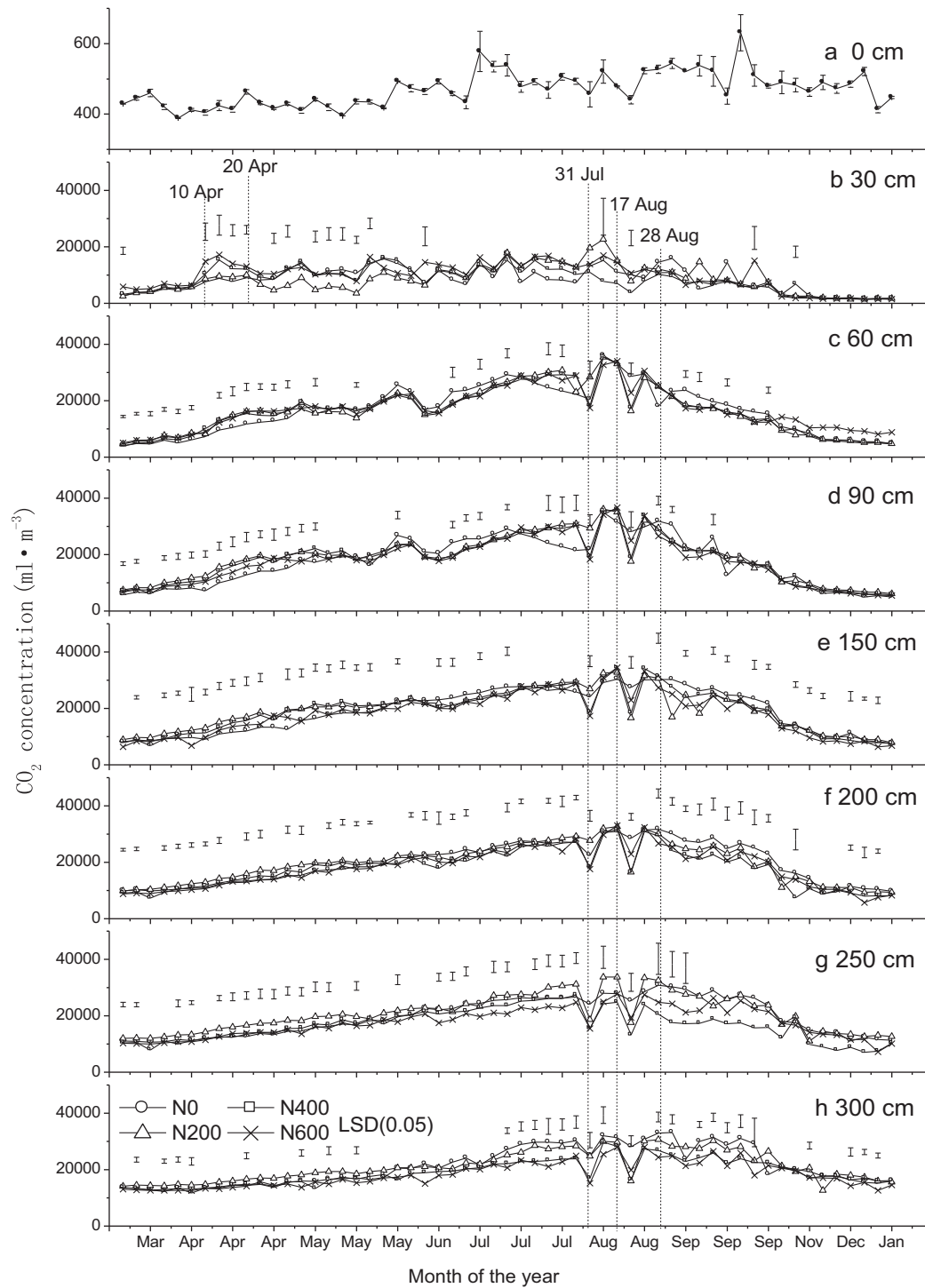
Fig. 6. Carbon dioxide concentrations in soil air (in ml per m^3) at various soil depths in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha^{-1} year^{-1}, in 2007–2008. Bars in figures indicate 1 standard deviation (n = 3).

Temporary increases in concentrations were most pronounced for N$_2$O.

Fertilizer N applications greatly increased grain and straw yields relative to the control (Table 4), but did neither affect the shape of the CH$_4$, CO$_2$ and N$_2$O concentration profiles nor the height of the concentrations of CH$_4$ and CO$_2$. A summary of the mean and median concentrations of CH$_4$, CO$_2$ and N$_2$O for all soil depths and fertilizer application treatments is shown in Table 5. Fertilizer N applications increased the size of the N$_2$O peaks significantly, and mean N$_2$O

concentrations roughly increased with fertilizer N application rate (Table 5C, Figs. 4 and 7).

Concentration peaks and drops occurred at all soil depths nearly instantaneously, suggesting a relatively large soil diffusivity. We found no evidence for a significant source of either CH$_4$ or CO$_2$ or N$_2$O in the subsoil. Concentrations in the subsoil largely reflect sources of CO$_2$ and N$_2$O production in the topsoil, while atmospheric CH$_4$ was the main source of the CH$_4$ in the soil profile. Changes with depth were most pronounced in the top 60 cm.
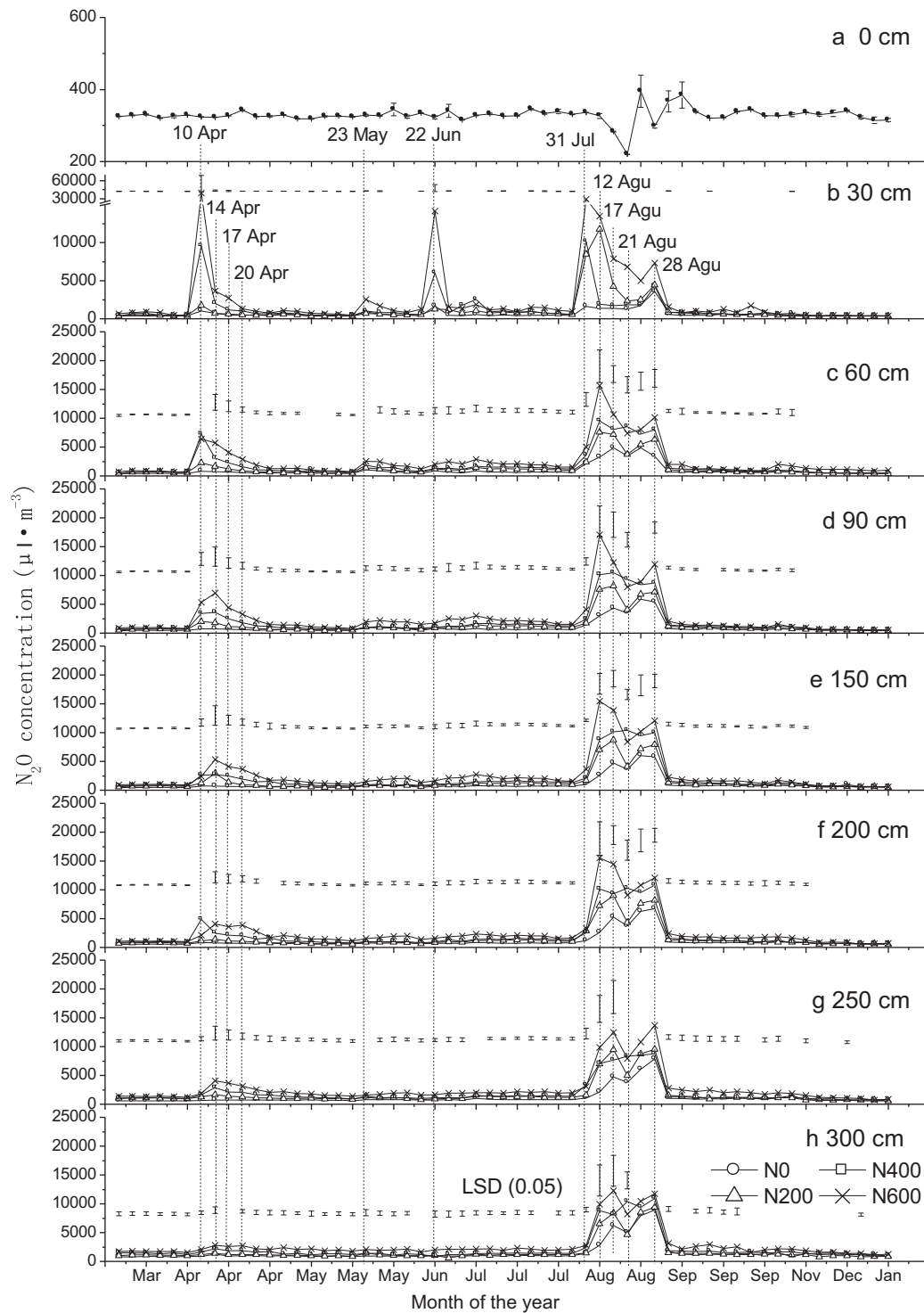
Fig. 7. Nitrous oxide concentrations in soil air (in μl per m³) at various soil depths in wheat–maize double cropping rotation receiving 0, 200, 400 and 600 kg of N ha⁻¹ year⁻¹, in 2007–2008. Bars in figures indicate 1 standard deviation (n = 3).

4.2. CH₄ concentration profiles

The magnitude of methane uptake by soils is largely controlled by diffusion of atmospheric CH_4 into the soil (Koschorreck and Conrad, 1993; Butterbach-Bahl, 2002), which in turn is strongly influenced by soil moisture (Striegl et al., 1992). Concentrations decreased with soil depth, suggesting oxidation of CH_4 by methanotrophic microorganisms. Between 60 and 300 cm, CH_4 concentrations remained more or less constant, suggesting that the methanotrophs were not able to take up and oxidize CH_4 at this low concentration. Our results suggests that CH_4 oxidation is strongest in the upper soil horizons, which is in agreement with observations made elsewhere (e.g. Hansen et al., 1993; Sitaula et al., 2000; Butterbach-Bahl, 2002; Wu et al., 2010).

CH_4 oxidation rate is negatively related to soil water content, primarily due to the control of the water filled pore space on the diffusivity of CH_4 through the soil profile, and the activity of methanotrophs (Nesbit and Breitenbeck, 1992; Dunfield

et al., 1995; Castro et al., 1995; Dobbie and Smith, 1996; Gulledge and Schimel, 1998). After heavy rainfall and irrigation, the WFPS increased (Fig. 2, from 10 to 20 April; 19 to 25 May and June to July), and the topsoil CH_4 concentration increased concomitantly (Fig. 5, from 10 to 20 April; 19 to 25 May and June to July). In the subsoil (from 60 to 90 cm), a high soil water content also coincided with a relatively high soil CH_4 concentration (see Figs. 2c,d and 5c,d). The CH_4 production increased with increasing temperature (Bergman et al., 1998), which could partially explain why CH_4 emissions increased quickly in maize season than that in wheat season at 30 cm (Table 3, Fig. 5).

Urea application did have a statistical significant effect on CH_4 concentrations, in the topsoil and subsoil. It has been suggested that fertilizer N application decreases CH_4 consumption (Hütsch et al., 1994; Mosier et al., 1998), either by an immediate inhibition of methanotrophs or by a change in microbial community due to repeated fertilizer N application. However, our results do not confirm such a relationship. Mean concentrations of CH_4 tended to be higher in the unfertilized control (see Fig. 5c and d), suggesting that N fertilizer application enhanced CH_4 oxidation. We relate this apparent anomaly to the higher WFPS of the control (Fig. 2c and d) and the lower dry matter production (Table 4).

4.3. CO_2 concentration profiles

The seasonal pattern of changes in CO_2 concentrations in the soil profile correlated with the soil temperature envelope, and has been attributed to CO_2 respiration following the rapid growth of plant root biomass during the growing season (Drury et al., 1998; Ruser et al., 2006; Bertora et al., 2008).

The seasonal cycle is affected by N fertilizer application, irrigation and heavy rainfall events. Heavy rainfall, N fertilizer application and irrigation events were succeeded by temporary increases in CO_2 concentrations, at basically all soil depths (see Figs. 2 and 6, from 10 to 20 April and the whole of August), but most in the topsoil. Evidently, heterotrophic respiration was most intense in the upper soil layers, where microbial activity is stimulated by organic carbon substrates supplied from the roots in the form of exudates and dead cells (Baudoin et al., 2001). Readily decomposable organic matter and ammonium have been shown to result in stimulation of microbial activity as evidenced by increased basal respiration.

Soil temperature, soil water content and drying/re-wetting cycles may have pronounced influences on soil CO_2 effluxes (Priemé and Christensen, 2001; Borken and Matzner, 2009). In our study area, the summer season is warm and relatively wet, while winter is cold and dry. This pattern is reflected in the soil temperatures measured between 30 and 150 cm in the studied profiles, which averaged 10 and 21 °C for the winter-wheat and summer-maize seasons, respectively (Table 3, Fig. 3). Within a certain temperature range, the CO_2 evolution from soil due to microbial respiration is directly correlated with soil temperature (Bajracharya et al., 2000). A positive relationship between soil temperature and biotic CO_2 production is commonly observed in soils (Schimel et al., 1994). Rates of CO_2 production were considerably higher during the warm maize growing season than during the cold wheat growing season (Tables 3 and 5B, Fig. 6). In general, the upper soil horizons (0–60 cm) are very dry in the dry season (before the middle of April and after the middle of November) (Fig. 2), which severely reduces biotic CO_2 production. In contrast, subsurface layers (>60 cm) retain higher levels of soil moisture throughout the dry season (Fig. 2), which sustains low, but continuous biotic CO_2 production (Fig. 6). CO_2 concentrations were positively related to soil water contents (Fierer et al., 2005), an increase in soil water content in the topsoil increases soil CO_2 concentrations by simultaneously increasing the rates of biotic CO_2 production and reducing the efflux

of CO_2 from the soil. A similar annual pattern in surface-versus-subsurface CO_2 production has been observed in other semiarid ecosystems (Wood and Petraitis, 1984; Keller and Bacon, 1998). In our study, CO_2 concentrations tended to increase with soil depth (Fig. 4, Table 5B), which is a common observation (e.g. Fierer et al., 2005).

4.4. N_2O concentration profiles

Concentrations of N_2O in the soil air responded rapidly and drastically to N fertilizer applications and irrigation events. Similar bursts of N_2O in soil after application of N fertilizer have been reported before (e.g. Ruser et al., 2001; Scheer et al., 2008; Van Groenigen et al., 2005; Reth et al., 2008; Liang et al., 2009). Concentration peaks were highest in the top 30 cm (Fig. 7) and lasted for one to several weeks. Peaks faded away at nearly the same sampling date, for all soil depths. Based on the appearance, disappearance and the size of the peaks, we speculate that most of the N_2O was produced in the surface 30 cm and from there diffused to the subsoil and the atmosphere. However, we have no concurrent N_2O production measurements and surface flux measurements to support this.

Highest N_2O concentrations were observed immediately following N fertilizer application and irrigation, in all soil layers (Fig. 7). Nitrogen application is a main trigger for N_2O production (Li et al., 1997; Jørgensen et al., 1998; Matson et al., 1998; Ruser et al., 2001; Scheer et al., 2008). Peaks of N_2O in the 30 and 60 cm horizons appeared on day 3 after application; in the other five horizons, the peaks appeared on day 7 after application, in all treatments except the control (Fig. 7). Such slight delays in the appearance of peak concentrations in the subsoil have also been reported by Scholefield et al. (1997) and Jahangir et al. (2012). The rapid response of the N_2O production in the topsoil horizon after N fertilizer application and irrigation is probably related to the presence of active soil microorganisms, which feed on the abundant sources of organic matter and nitrate (see Table 1). The slight delay in the appearance of N_2O peaks in the subsoil is probably related to (i) diffusion of N_2O from the topsoil to the subsoil, (ii) transport of substrates (nitrate/nitrite, dissolved organic carbon) to the subsoil, and (iii) the smaller population of active soil microorganism in the subsoil, compared to the topsoil. Our measurements do not allow to discriminate between these three processes, but the first process seems to have been most important for developing the concentration profiles shown in Fig. 7. This assertion is supported by results presented by Weitz et al. (2001), Chatskikh et al. (2005) and Grandy and Robertson (2006).

The soil water content has a dominant effect on the net N_2O efflux (e.g. Ruser et al., 2001; Metay et al., 2011), but also on the relative importance of nitrification and denitrification processes as sources of N_2O (e.g. Wrage et al., 2001). Zou et al. (2001) reported that nitrification is the predominant process contributing to N_2O fluxes during the maize growing season in dry land soil of Northern China. In maize season, from late June to the end of July (before applying nitrogen) no obvious diurnal pattern in soil N_2O production was observed in each horizon (see Fig. 7). Highest N_2O production through nitrification have been reported to occur between 30 and 70% WFPS, whereas highest N_2O fluxes by denitrification occur between 50 and 90% WFPS with a maximum at 70% (Bateman and Baggs, 2005; Gilliam et al., 2010). According to Davidson and Swank (1990) and Davidson (1992) N_2 starts being emitted through denitrification at a WFPS of 70%, and is the main N gas emitted when WFPS exceeds 75%; this may explain the weakness of simulated N_2O production from late June to the end of July (before applying nitrogen) (see Fig. 2). A double N_2O production peak emerged from 31 July to 28 August after N fertilizer application and irrigation; and the demarcation

point of the double peak occurred exactly on 21 August (see Fig. 7). The WFPS values were lower than 70% from 21 August (see Fig. 2), suggesting that nitrification is likely the predominant process contributing to N_2O production from 21 to 28 August. Since the temporal N_2O production stimulated by addition of N usually lasts only 1–3 weeks (Zou et al., 2001; Grandy and Robertson, 2006); we assumed the proper soil moisture conditions directly resulted in the N_2O production peak from 21 to 28 August. At our study site, WFPS varied between 45 and 70%, suggesting indeed that nitrification is likely a main source of the N_2O in the soil profile (Fig. 2). However, our measurements do not allow to conclude which microbiological pathway was the dominant source of N_2O.

Soil temperature influences nitrification rate and the rate of N_2O released per unit of ammonium-N oxidized (Goodroad and Keeney, 1984; Reth et al., 2005; Ruser et al., 2006; Lang et al., 2010, 2011). Mean soil temperatures in the topsoil were 10 and 21 °C for the wheat and maize seasons, respectively (see Fig. 3, Table 3). Peak N_2O concentrations were indeed higher in the summer season than in the winter season, but our data do not allow to conclude that the relative importance of nitrification and denitrification as source of N_2O was related to soil temperature.

5. Conclusions

Concentrations of CO_2, CH_4 and N_2O in soil air of 3 m deep soil profiles were determined at four fertilizer N application rates (0, 200, 400 and 600 kg N ha^{-1} year^{-1}) over a full year in a long-term field experiment with an irrigated winter wheat–summer maize double cropping system. Concentrations of CH_4 decreased with soil depth and showed little or no seasonal pattern, indicating that diffusive transport and consumption processes shaped the seasonal changes in concentration profiles. In contrast, concentrations of CO_2 increased with soil depth and showed a clear seasonal cycle, indicating that steady, seasonal-related CO_2 production processes and diffusive transport shaped the profiles in soil. Concentrations of N_2O also increased with depth, but patterns were predominantly related to incidental management activities, boosting short-term production processes which exceed N_2O consumption (i.e. reduction to N_2).

Urea application did not have a large effect on CH_4 concentrations, neither in the topsoil nor the subsoil, though mean concentrations were slightly higher in the unfertilized control than in the N fertilized treatments, likely because of the differences in WFPS due to differences in crop yield. Urea application also did not affect CO_2 concentrations significantly. However, it significantly increased mean N_2O concentrations peaks at basically all soil depths. Interestingly, concentrations of N_2O increased almost instantaneously in the whole soil profile, which indicates that the soil had a relatively high diffusivity, despite compacted subsoil layers.

The application of 200 kg urea N per ha per year tripled the sum of wheat and maize grain yields relative to the control, while mean N_2O concentrations were not more than doubled. This finding supports the finding of Van Groenigen et al. (2010) that the 'yield-scaled' N_2O emission (or yield scaled N_2O concentrations) is lowest at near optimal N application rates.

Changes in the concentrations of N_2O, CO_2 and CH_4 in soil were mainly related to the climatic factors temperature and water, and to the management factors N fertilizer application and irrigation. Changes were most profound in the topsoil. The subsoil was neither a significant source nor a significant sink of N_2O, CO_2 and CH_4; it acted largely as buffer.

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