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Annual methane uptake from different land uses in an agro-pastoral ecotone of northern China

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ABSTRACT

The semi-arid grasslands in northern China are subjected to accelerating land use change due to population growth and food demand. Considerable uncertainty exists on annual methane (CH_4) fluxes of different land uses because most measurements have only been conducted during the growing season. Using static chamber – gas chromatographic technique, we quantified and characterized annual CH_4 fluxes (from 2012 to 2015) from four land uses common in an agro-pastoral ecotone of northern China: summer-grazed grassland (SumGrazed), winter-grazed grassland (WinGrazed), ungrazed grassland since 1997 (Ungrazed) and oat cropland (OatCrop). The soil at all land uses functioned exclusively as a sink for atmospheric CH_4 through the entire three years. Annual CH_4 uptake rates averaged 1.42, 2.36, 1.12 and 2.57 kg C ha^{-1} yr^{-1} for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively, during 2012–2015. Compared to Ungrazed, OatCrop and WinGrazed increased annual CH_4 uptake by 129 and 111%, respectively. Non-growing season (October–April) contributed 28–43% of the annual CH_4 uptake at all land uses. Across all four land uses, annual cumulative CH_4 uptake decreased with increasing soil water-filled pore space (WFPS) explaining 81% of the variance in annual CH_4 uptake. WFPS negatively correlated to CH_4 uptake in the growing season (R^2 = 0.16-0.35, P < 0.001). CH_4 uptake increased with soil temperature through the entire observed period (R^2 = 0.38-0.63, P < 0.001) and the non-growing season (R^2 = 0.51–0.74, P < 0.001). We conclude that grazing has the potential to increase CH_4 uptake from the atmosphere and consequently, contribute positively to CH_4 related part of C budget.

1. Introduction

Next to carbon dioxide (CO_2), methane (CH_4) is the most important greenhouse gas with a global warming potential 34 times that of CO_2 (IPCC, 2013). Atmospheric CH_4 concentration has increased from 722 ppb to 1803 ppb since preindustrial times (IPCC, 2013). The predominant CH_4 sink is the reaction of CH_4 with hydroxyl radical in the troposphere, aerobic soil is the second largest global sink for atmospheric CH_4, estimated to be of 28–32 Tg yr^{-1} (IPCC, 2013).

Consumption of atmospheric CH_4 by soils is mainly controlled by the activity of methanotrophic bacteria, which can be strongly affected by soil disturbance, such as land-use change and agricultural practices (Ojima et al., 1993; Le Mer and Roger, 2001). Land-use change can greatly alter the CH_4 consumption by changing soil chemical and physical properties as well as microbial activities. Many studies indicated that cropland absorbed less CH_4 than grassland due to tillage and nitrogen fertilization (Mosier et al., 1991; Hütsch, 2001; Ding et al., 2004). However Rong et al. (2015) reported that cropland had higher CH_4 uptake than grassland because of the decrease in soil bulk density. Grazing has been reported to have both positive (Chen et al., 2011; Rong et al., 2015) and negative effects (Mosier et al., 1991; Liu et al., 2007; Holst et al., 2008) on CH_4 uptake. These varied results might be related to site-

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specific differences in grazing management, climate, soil type and properties.

Temperate grasslands, which currently account for about 10% of the Earth's surface, are very important to balance the budget of atmospheric CH_4 (White et al., 2000). Current estimates for annual CH_4 uptake of temperate grassland ecosystems are still highly uncertain, especially in Eurasian regions, because most measurements have only been conducted during the growing season (Liu et al., 2007; Wei et al., 2012; Tang et al., 2013). However, CH_4 oxidation still occurs despite soil temperature is below freezing point (Mosier et al., 1996; Wang et al., 2005; Chen et al., 2010, 2011). Since non-growing season lasts for more than half of the year in many temperate grassland regions, CH_4 consumption in the non-growing season may significantly contribute to annual uptake. A few field measurements showed that non-growing season CH_4 uptake accounted for 23–59% of annual CH_4 uptake (Chen et al., 2010, 2011; Rong et al., 2015). So, the CH_4 uptake in non-growing season is far from being ignored in the annual budget.

The semi-arid temperate grasslands in northern China account for about 78% of the national grassland area (Chen and Wang, 2000). Overgrazing and cultivation due to population growth and food demand are major causes of grassland degradation and desertification in these areas. Currently, complete and periodical exclusions of grazing have been used widely as approaches for restoring grasslands (Yang et al., 2015). A few field studies have addressed the effects of grazing management on CH_4 fluxes in the grasslands in northern China (Wang et al., 2005; Liu et al., 2007; Chen et al., 2010, 2011). However, influences of conversion from grasslands to croplands on CH_4 fluxes have received much less attention. Furthermore, most of these data are short-term (e.g. growing season or ≤2 year) and do not reflect seasonal and interannual variability of CH_4 fluxes. Therefore, long-term effects of land use change on soil CH_4 consumption in the semi-arid grasslands still remain unclear and need further research.

In present study, a three-year field study was carried out to estimate annual CH_4 fluxes from four land uses at typical semi-arid temperate grasslands in the agro-pastoral zone of northern China. The influences of soil temperature and moisture on CH_4 fluxes were also investigated. Our hypotheses were: 1) different land use types would affect CH_4 uptake capacity, and 2) CH_4 consumption in the non-growing season may significantly contribute to annual uptake. The results of this study will improve understanding of atmospheric CH_4 uptake in semi-arid grasslands and help to identify the optimum land uses to increase CH_4 sink.

2. Materials and methods

2.1. Site description

The experimental site (National Grassland Ecosystem Observation and Research Station) lies in the typical agro-pastoral ecotone in the Guyuan county, Hebei province, Northern China (41°46′N, 115°40′E, 1460 m above sea level). The region has a semi-arid and temperate continental monsoon climate, characterized by a short growing season and long cold winter with a frost-free period of 80–110 days. The annual mean temperature (1979–2009) is 1.4 °C with the maximum mean of 17.6 °C in July and the minimum of −18.6 °C in January. The annual mean precipitation is 380 mm with 80% occurring between May and September.

Four land use types common in this area were tested: a summer-grazed grassland (SumGrazed), a winter-grazed grassland (WinGrazed), an ungrazed grassland (Ungrazed), and a cropland (OatCrop). The OatCrop, Ungrazed and SumGrazed sites were adjacent to each other and the WinGrazed site lay about 1800 m west of these sites. The SumGrazed (15 ha) has been grazed at an intensity of 4–5 sheep ha^{-1} from June to September since 2009. The WinGrazed (10 ha) has been grazed by 5–6 sheep ha^{-1} from November to March since 2007. The Ungrazed (3 ha) was fenced in 1997 to forbid grazing. Grazing management changed vegetation composition and coverage (Table 1). The cropland (5 ha) was converted from native grassland to cropland in 1995 and Oat (*Avena sativa*) monoculture has been grown annually since then. In late May, just before sowing Oat, the cropland was ploughed and turned to 20 cm depth. Crop residue was removed after harvest (in late September) and only 5–10 cm of stubble remained. Neither irrigation nor herbicides were applied to any of the land uses and only 675 kg solid cattle manure ha^{-1} (corresponding to 30 kg N ha^{-1}) was applied to the cropland as the base fertilizer before sowing. Soils for all four land uses are classified as Kastanozems according to the FAO system. The main characteristics of the four land uses are shown in Table 1 (Yang et al., 2015).

2.2. CH_4 flux measurements

At each land use, four replicate chambers were randomly designated to measure CH_4 fluxes applying static opaque chamber and gas chromatography method (Yang et al., 2015). Chambers were spaced approximately 15 m apart. The sampling chamber was made of stainless steel consisting of two parts: a square base frame (0.5 m × 0.5 m, height 0.1 m) and a removable top lid (0.5 m × 0.5 m, height 0.5 m) on which had a gasket to ensure air-tightness. The white heat-insulating foam was added outside of top chamber to reduce the impact of direct radiative heating during sampling. There was a hole located on the top wall of the upper chamber. The hole was connected with a 12-cm long silicon tube (1 cm in diameter) to minimize pressure perturbations when collecting air samples. Methane fluxes were measured with the aggressive sampling scheme during three years: from 30 May 2012 to 30 May 2015. Measurements were obtained twice per week during growing season (May–September, the duration from grass emergence to senescence) and during spring thaw (March–April), and twice per month from October to February due to harsh climatic conditions. The gas samplings were always performed between 8:00–11:00 to represent daily average flux (Yang et al., 2015) with 60-ml plastic stopcock-syringes. Chambers were closed for one hour and five gas samples were collected at 15 min intervals. CH_4 concentrations of the gas samples stored in syringes were measured within 8 h after sampling with a flame ionization detector (FID) in a gas chromatograph (Agilent 7890A, Santa Clara, CA, USA). CH_4 flux was calculated jointly from the linear (Eqs. (1) and (2)) or non-linear change in gas concentrations (Eqs. (1) and (3)) (Kroon et al., 2010; Wang et al., 2013). Daily CH_4 fluxes were estimated by linear interpolation for non-sampling days. Seasonal and annual cumulative CH_4 uptake was summed from the measured and interpolated values (Chen et al., 2011).

$$F_{CH4} = dc/dt * \rho * H \tag{1}$$

$$C = a * t + b(dc/dt = a) \tag{2}$$

$$C = k_1/k_2 + (C_0 - k_1/k_2) * \exp(-k_2 * t)(dc/dt|_{t=0} = k1 - k2 * C0) \tag{3}$$

Where F_{CH4} is the hourly CH_4 uptake (μg C m^{-2} h^{-1}); ρ is the CH_4 gas density (μg m^{-3}); dc/dt is the rate of change in gas concentration inside the chamber (h^{-1}); H is the height of the chamber (m); C is the measured CH_4 concentration; C_0 is the concentration at the beginning of the enclosure; a, b, k_1 and k_2 are parameters derived by fitting linear or nonlinear curves.

Table 1
Main characteristics of studied land uses.

Land use	SumGrazed	WinGrazed	Ungrazed	OatCrop
pH	8.1 ± 0.1	7.7 ± 0.0	7.6 ± 0.0	7.9 ± 0.1
Bulk density ($g\,m^{-3}$)	1.11 ± 0.02	1.23 ± 0.08	1.45 ± 0.01	1.25 ± 0.01
SOC ($g\,kg^{-1}$)	28.2 ± 3.0	23.2 ± 1.6	13.0 ± 2.5	14.7 ± 1.3
Total N ($g\,kg^{-1}$)	3.2 ± 0.3	2.7 ± 0.2	1.5 ± 0.3	1.6 ± 0.2
Sand (%)	45.3 ± 1.1	50.6 ± 6.7	71.9 ± 1.7	61.0 ± 8.0
Silt (%)	35.9 ± 3.9	27.6 ± 7.6	17.7 ± 1.0	22.9 ± 5.2
Clay (%)	18.8 ± 3.1	21.8 ± 2.0	10.4 ± 1.0	16.1 ± 2.9
Soil texture	Loam	Sandy loam	Sandy loam	Sandy loam
Above aground biomass ($g\,m^{-2}$)	56 ± 12	307 ± 66	426 ± 82	336 ± 42
Root biomass ($g\,m^{-2}$)	1392 ± 142	1277 ± 188	1933 ± 370	737 ± 70
Vegetation composition	*Leymus chinensis* (73%), *Stipa krylivii* (10%), *Potentilla acaulis* (8%), *Iris lactea Var. chinesis* and *Saussurea runcinata* (9%)	*Leymus chinensis* (80%), *Stipa krylivii* (12%), *Artemisia frigida* and *Potentilla acaulis* (8%)	*Leymus chinensis* (90%), *Stipa krylivii* (8%), *Hordeum brevisubulatum* and *Potentilla acaulis* (2%).	Oat (*Avena sativa*)
Vegetation coverage	45%	60%	90%	N. D.

Data adapted from Yang et al. (2015).
Values are mean ± S.D (standard deviation), N.D.: not determined, SumGrazed: summer-grazed grassland; WinGrazed: winter-grazed grassland; Ungrazed: ungrazed grassland; OatCrop: cropland.

2.3. Auxiliary measurements

Soil temperature and moisture were measured in close vicinity to the chambers at the same time as each gas sampling. Soil temperature (at 5 cm soil depth) as well as volumetric water content (0–6 cm) were measured with portable digital thermometers (JM624, JinMing Instrument Co. Ltd., Tianjin, China) or portable frequency domain reflectometry probes (FDR, ThetaKit, Delta-T Devices, Cambridge, UK), respectively. Soil moisture (0–5 cm) was determined gravimetrically when the soil was frozen. Soil moisture was converted to percent water-filled pore space (%WFPS) according to Yang et al. (2015).

The relationship between CH_4 uptake flux and soil temperature was expressed as an exponential function (Chen et al., 2014):

$$CH_4 = a\exp(b * T) \tag{4}$$

Where a and b are the empirical coefficients, and T is the soil temperature at 5 cm depth in this study.

The Q_{10} value can be calculated as

$$Q_{10} = \exp(10 * b) \tag{5}$$

Where b is the value from Eq. (4)

2.4. Statistical analyses

One-way ANOVA was used to test differences in seasonal and annual CH_4 uptake among land uses. Linear or non-linear regressions were used to examine the relationship between CH_4 fluxes with soil temperature and WFPS. All statistical analysis was performed with SPSS 11.0 (SPSS Inc., Chicago, USA). Differences were considered significant at $P < 0.05$.

3. Results

3.1. Precipitation, air and soil temperature

Air temperature presented clear seasonal courses (Figs. 1a, 2a, 3a and 4a), being highest in July-August and lowest in January–February. Over the three-year period, annual mean air temperature differed (−1.8, 3.3 and 3.5 °C for 2012–2013, 2013–2014 and 2014–2015, respectively) from the 30-year average of 1.4 °C. Precipitation was lower than the 30-year average of 380 mm in the year 2012–2013 (297 mm) and 2014–2015 (335 mm) but higher in the year 2013–2014 (450 mm) (Figs. 1b, 2b, 3b and 4b). Precipitation during the growing season accounted for 83, 89 and 83% of total rainfall in 2012–2013, 2013–2014 and 2014–2015, respectively.

The seasonality of soil temperature coincided with the seasonal patterns of air temperature (Figs. 1a, 2a, 3a and 4a). Daily soil temperature varied widely within a range from −12.3 °C in February 2013 to 28.0 °C in July 2012 across all four land uses. Soil was frozen from the middle of November to the early March each year. During the study period, mean soil temperatures were 10.6, 12.0, 6.6 and 8.2 °C for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively. Soil WFPS were intimately linked to rainfall (Figs. 1b, 2b, 3b and 4b). During the spring thaw (from the middle of March to the end of April), WFPS increased in all land uses. For the study years, mean soil WFPS were 41, 28, 44 and 30%, ranging from 9 to 73, 4 to 56, 10 to 80 and 7 to 60% for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively.

3.2. CH_4 uptake rates

The soil at all land uses functioned exclusively as a sink for atmospheric CH_4 (Figs. 1c, 2c, 3c and 4c). CH_4 uptake rates at all land uses revealed distinct patterns of seasonal changes during the entire observation periods of 2012–2015 with high rates during the growing season and low rates during the frozen period. The three-year mean daily CH_4 uptake rates were 0.49, 0.86, 0.37 and 0.90 $mg\,C\,m^{-2}\,d^{-1}$, ranging from 0.08 to 1.10, 0.07 to 1.93, 0.05 to 0.87 and 0.13 to 1.96 $mg\,C\,m^{-2}\,d^{-1}$ for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively.

CH_4 uptake rates increased with soil temperature during the spring-thaw periods in all land uses, but we did not observe a pulse in CH_4 uptake in any of the land uses.

3.3. Seasonal and annual CH_4 uptake

The mean annual CH_4 uptake within the 2012–2015 period was 1.42, 2.36, 1.12 and 2.57 $kg\,C\,ha^{-1}\,yr^{-1}$ for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively (Table 2). The growing-season CH_4 uptake dominated the total annual uptake and accounted for 57–72% of annual uptake across all sites. The non-growing season CH_4 uptake on average accounted for 38, 31, 41 and 35% of annual CH_4 uptake at SumGrazed, WinGrazed, Ungrazed and OatCrop sites, respectively.

Annual CH_4 uptake was highest in 2014–2015 and lowest in 2012–2013. However, interannual variations were low and varied at best by 29% (e.g. at Ungrazed site, 2012–2013: 0.98 $kg\,C\,ha^{-1}\,yr^{-1}$; 2014–2015: 1.26 $kg\,C\,ha^{-1}\,yr^{-1}$).

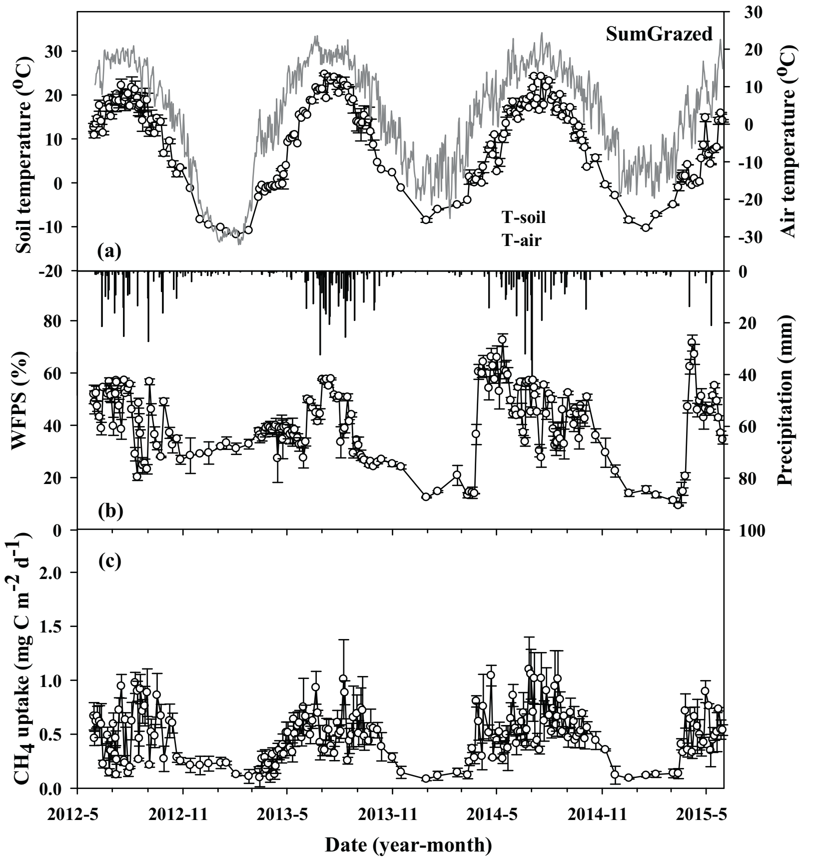

Fig 1. Seasonal variations of (a) soil temperature at 5 cm depth and air temperature, (b) water-filled pore space (%WFPS, 0–6 cm) and precipitation, (c) CH_4 uptake, at summer-grazed grassland (SumGrazed) from 30 May 2012 to 30 May 2015 in the agro-pastoral ectone of northern China. Values are mean ± S.E (standard error) (n = 4).

Table 2
Seasonal and annual cumulative CH_4 uptake depending on land uses from 30 May 2012 to 30 May 2015 in the agro-pastoral ectone of northern China.

Period	Land use	Cumulative CH_4 (kg C ha^{-1})			NGS contribution (%)
		Annual	GS	NGS	
2012/5–2013/5	SumGrazed	1.35 ± 0.13 b	0.84 ± 0.09 b	0.51 ± 0.04 b	38
	WinGrazed	2.26 ± 0.11 a	1.63 ± 0.08 a	0.63 ± 0.05 b	28
	Ungrazed	0.98 ± 0.06 c	0.61 ± 0.05 b	0.37 ± 0.02 c	38
	OatCrop	2.46 ± 0.08 a	1.59 ± 0.07 a	0.87 ± 0.06 a	35
2013/5–2014/5	SumGrazed	1.38 ± 0.09 b	0.83 ± 0.08 b	0.55 ± 0.01 c	39
	WinGrazed	2.28 ± 0.14 a	1.54 ± 0.09 a	0.74 ± 0.05 b	32
	Ungrazed	1.12 ± 0.04 b	0.66 ± 0.03 b	0.46 ± 0.02 c	41
	OatCrop	2.47 ± 0.12 a	1.52 ± 0.09 a	0.95 ± 0.08 a	38
2014/5–2015/5	SumGrazed	1.54 ± 0.11 b	0.95 ± 0.12 b	0.59 ± 0.02 b	38
	WinGrazed	2.53 ± 0.15 a	1.67 ± 0.10 a	0.86 ± 0.06 a	34
	Ungrazed	1.26 ± 0.03 b	0.72 ± 0.03 b	0.54 ± 0.05 b	43
	OatCrop	2.79 ± 0.08 a	1.89 ± 0.07 a	0.90 ± 0.02 a	32

Values are mean ± S.E (standard error). Values within a column followed by different lowercase letters are significantly different ($P < 0.05$), GS: growing season; NGS: non-growing season, SumGrazed: summer-grazed grassland; WinGrazed: winter-grazed grassland; Ungrazed: ungrazed grassland; OatCrop: cropland.

3.4. Effects of soil temperature and moisture on daily CH_4 uptake

For all land uses, daily CH_4 uptake increased with daily topsoil (5 cm) temperature (correlation through the entire observed period, $R^2 = 0.38–0.63$, $P < 0.001$, Fig. 5 and the non-growing season, $R^2 = 0.51–0.74$, $P < 0.001$, Fig. 6). However, in the growing season this correlation was not significant (Figure not shown). Soil temperature explained 38, 63, 48 and 61% of the variation of CH_4 fluxes over the entire study and 51, 74, 52 and 74% for the non-growing season for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively. Daily CH_4 uptake decreased with WFPS for all land uses during the growing season ($P < 0.001$, Fig. 7). Soil WFPS accounted for 23, 35, 16 and 24% of daily CH_4 uptake fluxes for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively.

4. Discussion

4.1. Annual CH_4 uptake

Soils at all land uses were efficient CH_4 sink through the entire three years. Seasonal changes in CH_4 oxidation were distinct (Figs. 1c, 2c, 3c and 4c) in accordance with observation of others (Wang et al., 2005; Chen et al., 2010, 2011). The annual CH_4 uptake from three grassland sites ranged from 0.98 to 2.53 kg C ha^{-1} yr^{-1}

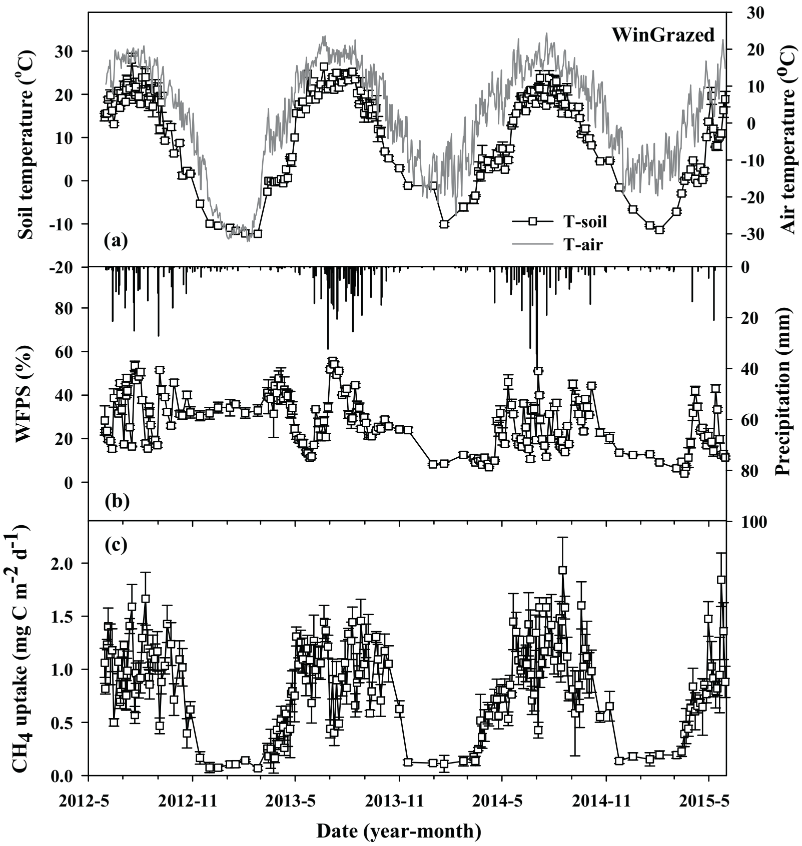

Fig. 2. Seasonal variations of (a) soil temperature at 5 cm depth and air temperature, (b) water-filled pore space (%WFPS, 0–6 cm) and precipitation, (c) CH_4 uptake, at winter-grazed grassland (WinGrazed) from 30 May 2012 to 30 May 2015 in the agro-pastoral ectone of northern China. Values are mean ± S.E (standard error) (n = 4).

(Table 2), which was in consistent with the report in the same area by Rong et al. (2015) (1.02–2.72 kg C ha^{-1} yr^{-1}). The CH_4 uptake for the WinGrazed site (2.26–2.53 kg C ha^{-1} yr^{-1}) was similar to the value of 2.3 kg C ha^{-1} yr^{-1} reported by Chen et al. (2011) in temperate semi-arid grassland in Inner Mongolia. The mean annual CH_4 uptake in our grasslands (1.63 kg C ha^{-1} yr^{-1}) was higher than those in humid grasslands, e.g. tropical grasslands in western Puerto Rico (0.40–0.59 kg C ha^{-1} yr^{-1}, Mosier and Delgado, 1997), grassland in Spain (0.9 kg C ha^{-1} yr^{-1}, Merino et al., 2004). The lower precipitation and better aerobic conditions in this temperate semi-arid region might favor CH_4 oxidation by methanotrophic bacteria and contribute to the enhanced soil CH_4 uptake.

The annual CH_4 uptake from OatCrop site ranged from 2.46 to 2.79 kg C ha^{-1} yr^{-1}. On average, the cropland soil consumed about 2.57 kg C ha^{-1} yr^{-1} from the atmosphere. This value was close to the value of 2.99 kg C ha^{-1} yr^{-1} obtained by Dobbie and Smith (1996) for their arable land in eastern Scotland. Our result was much higher than the uptake from rain-fed cropland in a semi-arid region of South-eastern Australia (0.60–1.08 kg C ha^{-1} yr^{-1}) (Barton et al., 2013). The lower CH_4 uptake was attributed to moisture limitations of the methanotrophic community during most of the observation period in South-eastern Australia (Del Grosso et al., 2000). The CH_4 uptake reported here was higher than the uptake from intensively fertilized cropland soils in the North China Plain (0.74–1.16 kg C ha^{-1} yr^{-1}, Hu et al., 2013; 1.12–1.54 kg C ha^{-1} yr^{-1}, Shi et al., 2013). The lower CH_4 uptake rates could be related to higher nitrogen application in the North China Plain, since fertilizer nitrogen applications have an inhibitory effect on atmospheric CH_4 uptake by soil (Mosier et al., 1991; Willison et al., 1996; Aronson and Helliker, 2010).

Annual CH_4 uptake at grazed sites (SumGrazed and WinGrazed) were on average 27% and 111% higher than those at Ungrazed, which might indicate that grazing increased consumption of atmospheric CH_4. The lower CH_4 uptake in the Ungrazed site was probably caused by a combination of higher WFPS and lower soil temperature compared to the SumGrazed and WinGrazed sites. Higher soil moisture can limit diffusion of CH_4 into the soil resulting in decreased CH_4 uptake (van den Pol-van Dasselaar et al., 1998). Lower soil temperature can depress CH_4 uptake. In the growing season, higher WFPS values in Ungrazed site (Ungrazed: 46%, SumGrazed: 44%, WinGrazed: 29%) were attributed to reduction in soil evaporation losses as a result of greater aboveground coverage. The shading by vegetation and litter in the Ungrazed site decreased the soil temperature during the growing season as compared to SumGrazed and WinGrazed sites (Ungrazed: 10.8 °C, SumGrazed: 16.2 °C, WinGrazed: 18.1 °C). In the non-growing season, the higher and denser standing vegetation biomass and more litter coverage in the Ungrazed site could increase snow accumulation. As a consequence, Ungrazed increased soil moisture as compared to the grazed sites (Ungrazed: 40%, SumGrazed: 36%, WinGrazed: 26%). While in the SumGrazed and WinGrazed sites, grazing removed most aboveground plant biomass and litter. SumGrazed and WinGrazed increased soil evaporation losses and reduced soil water content and increased soil temperature in the growing season. During the non-growing season, SumGrazed and WinGrazed weakened snow-holding capacity and enhanced soil drying by wind and sun, and consequently reduced soil water contents. Annual CH_4 uptake at WinGrazed site was 66% higher than that at SumGrazed, suggesting that WinGrazed was a better regime for enhancing CH_4 uptake. Lower CH_4 uptake at SumGrazed site may be due to the higher WFPS compared to WinGrazed. The

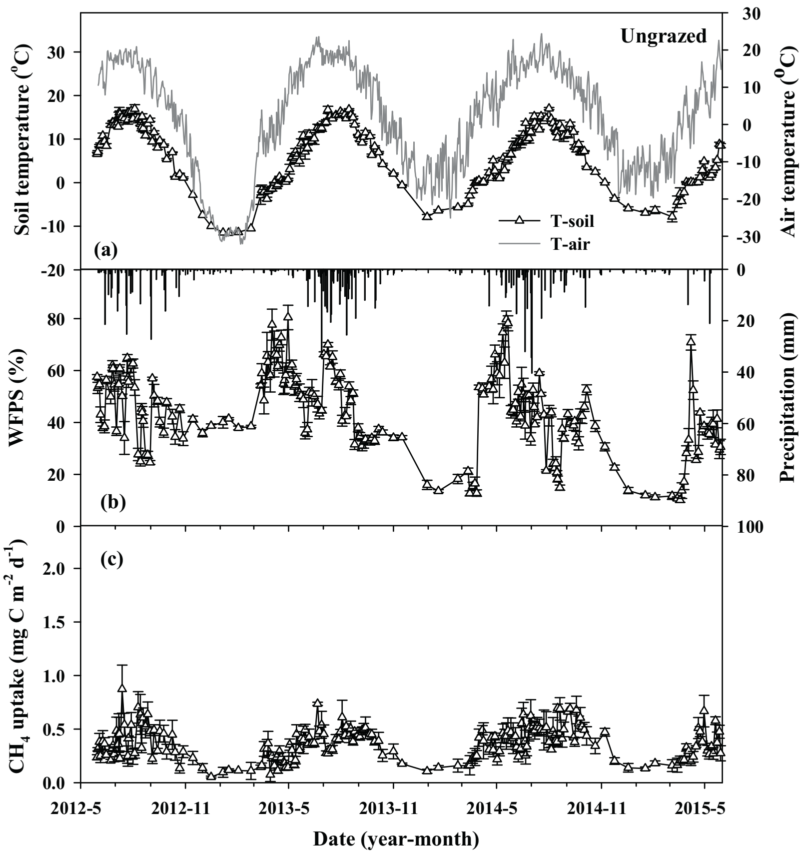

Fig. 3. Seasonal variations of (a) soil temperature at 5 cm depth and air temperature, (b) water-filled pore space (%WFPS, 0–6 cm) and precipitation, (c) CH_4 uptake, at ungrazed grassland (Ungrazed) from 30 May 2012 to 30 May 2015 in the agro-pastoral ectone of northern China. Values are mean ± S.E (standard error) (n = 4).

increased WFPS value was a result of increased soil water conservation at SumGrazed site due to finer texture compared to WinGrazed (Table 1). Our findings confirmed the results of Chen et al. (2011) and Rong et al. (2015) who reported that light (0.92 sheep ha^{-1} yr^{-1}) and moderate grazing (1.43 sheep $ha^{-1}yr^{-1}$) would promote CH_4 uptake in semi-arid temperate grasslands in north China. However, our result was in contrast to other findings (Mosier et al., 1991; Liu et al., 2007; Holst et al., 2008). These studies have shown that grazing reduced annual CH_4 uptake by (1) soil compaction as a result of animal treading which decreases the diffusion of CH_4 and O_2; (2) fresh animal feces deposition which are hotspot sources of CH_4; (3) increasing the likelihood of physiological water stress which could inhibit the activities of methanotrophs; and (4) changing the community of soil methanotrophs (Liu et al., 2007; Chen et al., 2011; Wang et al., 2012; Tang et al., 2013).

The three-year mean annual CH_4 uptake from cropland site were 9–129% higher than those from grassland sites, suggesting that land use change from grassland to cropping increased CH_4 uptake. Higher CH_4 uptake at OatCrop site was connected with (1) lower WFPS (30%) and (2) cattle manure addition. Smaller amounts of manure application (ca. 30 kg N ha^{-1}) may promote growth and activity of methanotrophs and accelerate methane oxidation. Our observations were consistent with those of Rong et al. (2015) who found higher annual CH_4 uptake in cropland than in grasslands. However, our result was in contrast to the reports that the conversion from forest or grassland to croplands significantly reduced oxidation of CH_4 (Mosier et al., 1991; Ojima et al., 1993; Hütsch, 2001; Ding et al., 2004). In these studies, the reduced CH_4 oxidation in agricultural soils was related to soil disturbance and N application. Disturbance for instance by ploughing may reduce the probability of the biological, chemical and physical parameters that define the ecological niche for methanotrophic bacteria occurring (Willison et al., 1995; Hütsch, 1998). On the other hand, disturbance may reduce CH_4 uptake through compaction because of the enhanced diffusion resistance for CH_4 and O_2. N application exhibited a strong inhibitory effect on CH_4 oxidation since ammonium may compete with CH_4 oxidation enzymes (Steudler et al., 1989). The factors, such as crop type, agricultural management, quantity and type of fertilization, climate and soil, need to be accounted for the diverse effects in different researches.

Rapid changes in CH_4 uptake during the transitions between the growing and non-growing seasons were evident in the SumGrazed, WinGrazed and OatCrop sites, but not in the Ungrazed site (Figs. 1–4). Both higher soil moisture (Ungrazed: 46%, SumGrazed: 44%, WinGrazed: 29%, OatCrop: 30%) and lower soil temperature (Ungrazed: 10.8 °C, SumGrazed: 16.2 °C, WinGrazed: 18.1 °C, OatCrop: 13.1 °C) mostly likely inhibited growing season CH_4 uptake in the Ungrazed site as compared to the other sites. The integrated effects of soil temperature and WFPS explained 30, 39, 24 and 29% of the variation of daily CH_4 uptake for SumGrazed, WinGrazed, Ungrazed and OatCrop, respectively (Fig. 8).

Across all land uses, both growing season and annual CH_4 uptake was negatively correlated with WFPS ($P < 0.001$, Fig. 9). Soil WFPS accounted for 88 and 81% of the variances in growing season and annual CH_4 uptake, indicating that the strength of both growing season and annual CH_4 uptake under land uses was largely determined by soil water contents.

4.2. CH_4 uptake during the non-growing season

Regardless of the land use or year, soils in the agro-pastoral ectone of northern China took up atmospheric CH_4 throughout

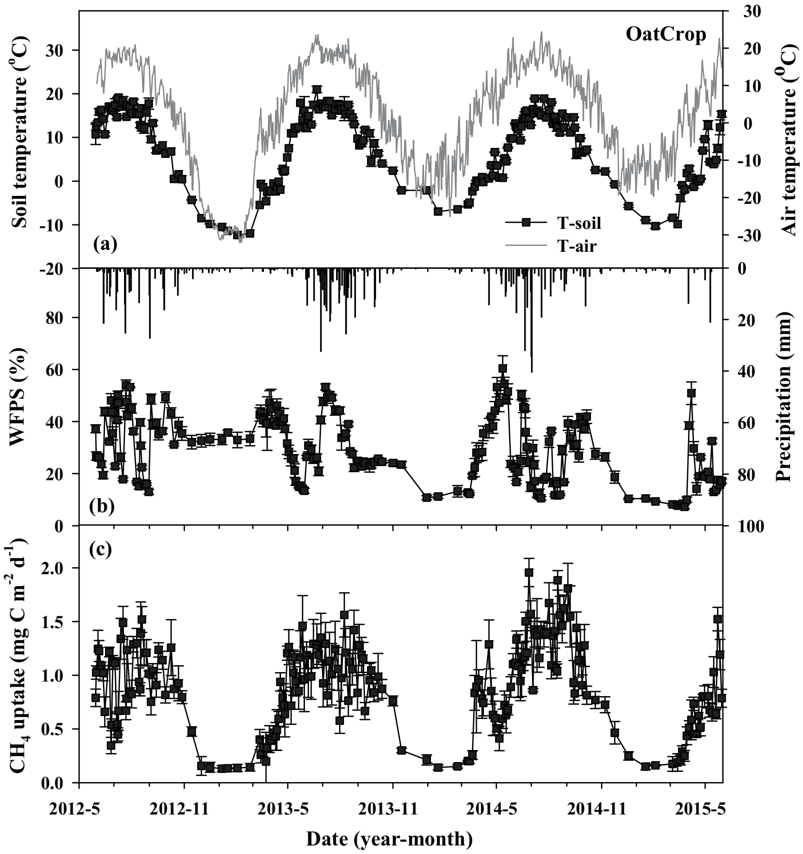

Fig. 4. Seasonal variations of (a) soil temperature at 5 cm depth and air temperature, (b) water-filled pore space (%WFPS, 0–6 cm) and precipitation, (c) CH_4 uptake, at oat cropland (OatCrop) from 30 May 2012 to 30 May 2015 in the agro-pastoral ectone of northern China. Values are mean ± S.E (standard error) (n = 4).

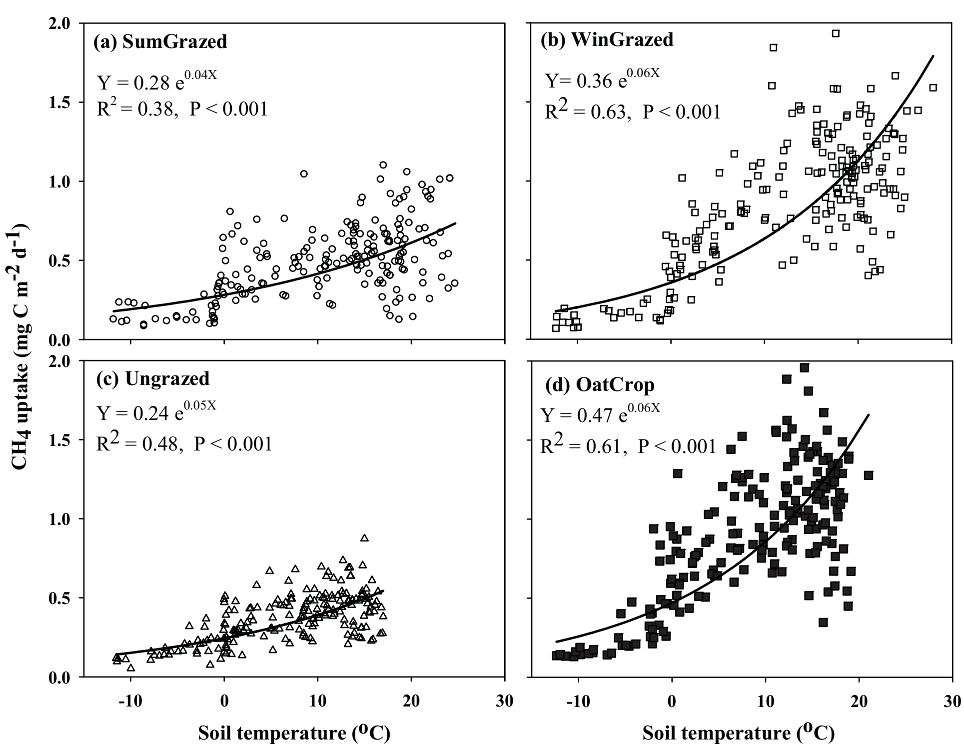

Fig. 5. Relationship between CH_4 uptake and soil temperature at 5 cm depth through the observation period depending on land uses: (a) summer-grazed grassland (SumGrazed), (b) winter-grazed grassland (WinGrazed), (c) ungrazed grassland (Ungrazed) and (d) cropland (OatCrop).

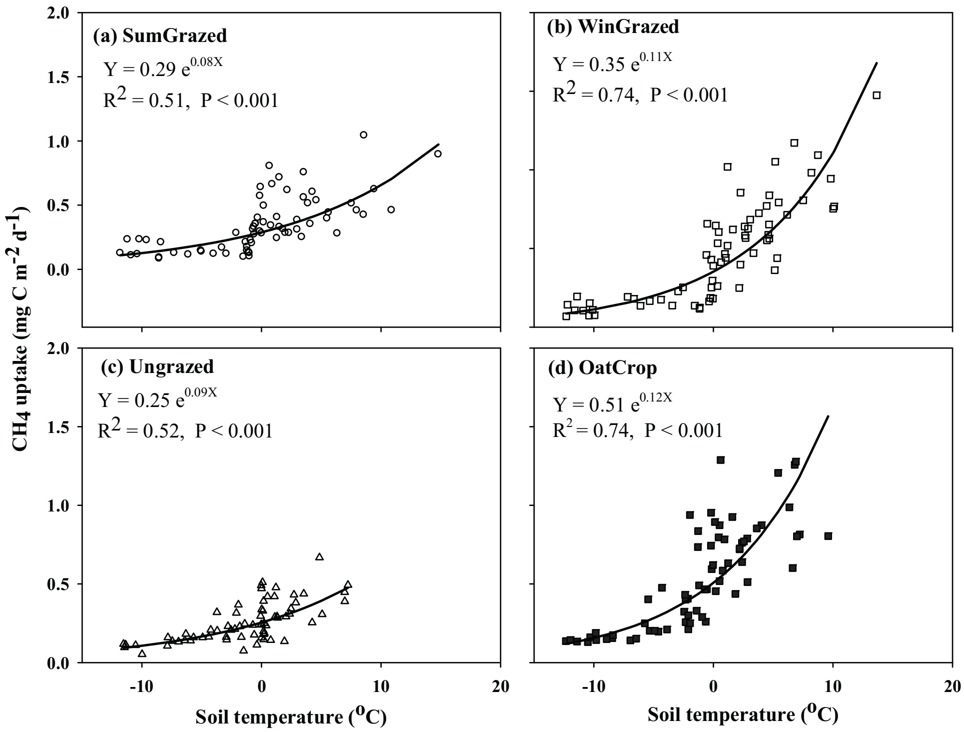

Fig. 6. Relationship between CH_4 uptake and soil temperature at 5 cm depth during the non-growing season depending on land uses: (a) summer-grazed grassland (SumGrazed), (b) winter-grazed grassland (WinGrazed), (c) ungrazed grassland (Ungrazed) and (d) cropland (OatCrop).

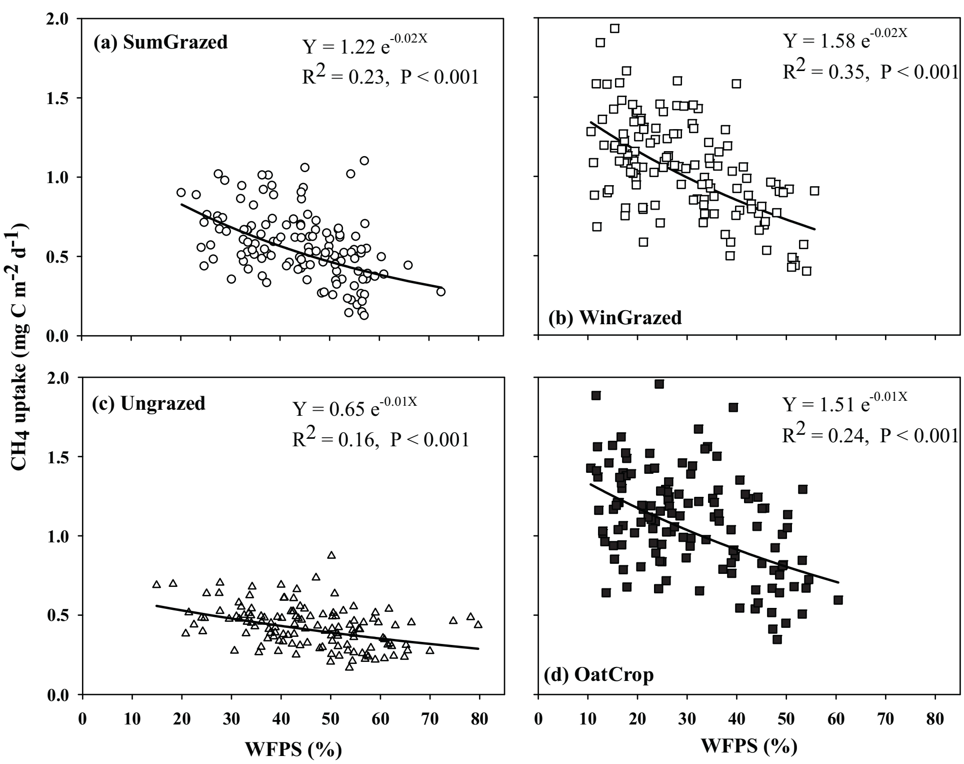

Fig. 7. Relationship between CH_4 uptake and water-filled pore space (%WFPS, 0–6 cm) during the growing season depending on land uses: (a) summer-grazed grassland (SumGrazed), (b) winter-grazed grassland (WinGrazed), (c) ungrazed grassland (Ungrazed) and (d) cropland (OatCrop).

the freezing period. A similar phenomenon has been observed in grasslands, forests and agriculture ecosystems (Mosier et al., 1996; Butterbach-Bahl and Papen, 2002; Dörsch et al., 2004; Chen et al., 2010, 2011). CH_4 oxidation during soil freezing has been attributed to: (1) microbiological activity in unfrozen soil water located in a thin water film at the soil matrix (Tamai et al., 2003); (2) a shift in CH_4 uptake activity to deeper, unfrozen soil horizons (Chen et al., 2010). The warmer conditions in the deeper horizon might facilitate

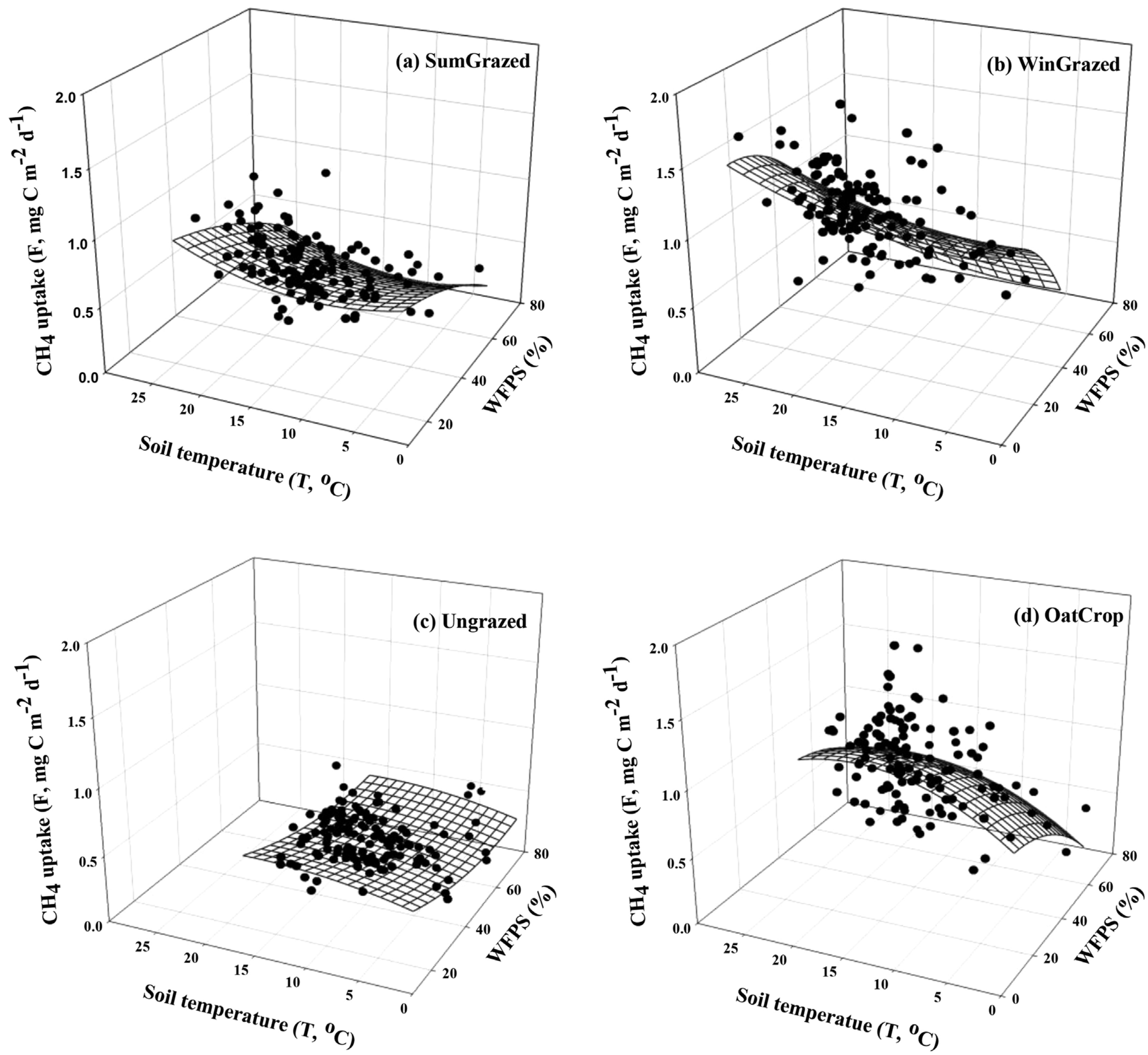

Fig. 8. Relationship between CH_4 uptake and water-filled pore space (%WFPS, 0–6 cm) and soil temperature (T, °C) during the growing season depending on land uses: (a) summer-grazed grassland (SumGrazed), (b) winter-grazed grassland (WinGrazed), (c) ungrazed grassland (Ungrazed) and (d) cropland (OatCrop).Fitting curves: (a) SumGrazed, $F = 0.88 - 1.16\,e^{-3} * WFPS - 0.02 * T - 1.01\,e^{-4} * (WFPS)^2 + 1.16\,e^{-3} * T^2$ ($R^2 = 0.30$, $P < 0.001$), (b) WinGrazed, $F = 1.07 + 8.65\,e^{-3} * WFPS - 7.50\,e^{-3} * T - 3.77\,e^{-4} * (WFPS)^2 + 6.79\,e^{-4} * T^2$ ($R^2 = 0.39$, $P < 0.001$), (c) Ungrazed, $F = 0.64 - 0.01 * WFPS + 0.02 * T + 8.19\,e^{-5} * (WFPS)^2 - 6.05\,e^{-4} * T^2$ ($R^2 = 0.24$, $P < 0.001$), (d) OatCrop, $F = 0.83 + 1.40\,e^{-3} * WFPS + 0.06 * T - 1.82\,e^{-4} * (WFPS)^2 - 1.84\,e^{-3} * T^2$ ($R^2 = 0.29$, $P < 0.001$).

CH_4 oxidation under unfavorable conditions near the soil surface (Striegl et al., 1992).

Non-growing season CH_4 uptake constituted 28–43% in the grassland soils and 32–38% of annual CH_4 uptake in the cropland soil (Table 2). Rong et al. (2015) observed that the contribution of the non-growing season to CH_4 uptake represented 22–59% for the grassland soils and 20% for the cropland soil in the same region. Chen et al. (2010, 2011) reported that the contributions of the non-growing season to the annual CH_4 uptake ranged from 23 to 40% for temperate semiarid grasslands in Inner Mongolia. Mosier et al. (1996) found that wintertime (from November to February) CH_4 consumption contributed 15–30% of the annual uptake for a semiarid prairie in Northern America, which was in agreement with our results (from October to the middle of March, 21–28%). These researches highlighted that the significant contribution of non-growing season to the annual CH_4 budge cannot be neglected

4.3. Effects of soil temperature and moisture on CH_4 uptake

Soil temperature is positively correlated with CH_4 uptake in many ecosystems (Castro et al., 1995; Butterbach-Bahl and Papen, 2002; Maljanen et al., 2003; Chen et al., 2010, 2011). The dependence of CH_4 uptake on soil temperature can be attributed to sensitivity of microbial activity to temperature as consumption of atmospheric CH_4 by soil is microbial mediated. CH_4 uptake correlated ($P < 0.001$) over the entire observed period or only in the non-growing seasons with soil temperature (Figs. 6 and 7). This suggested that the lower CH_4 uptake in the non-growing seasons in comparison with that in the growing seasons was because of the lower soil temperature. Our findings were in line with the reports by Castro et al. (1995) and Steinkamp et al. (2001) that soil temperature was an important controller at low temperatures, but had no effect on CH_4 uptake at higher temperatures. The three-year mean Q_{10} values were 1.5, 1.8, 1.6 and 1.8 for SumGrazed, WinGrazed,

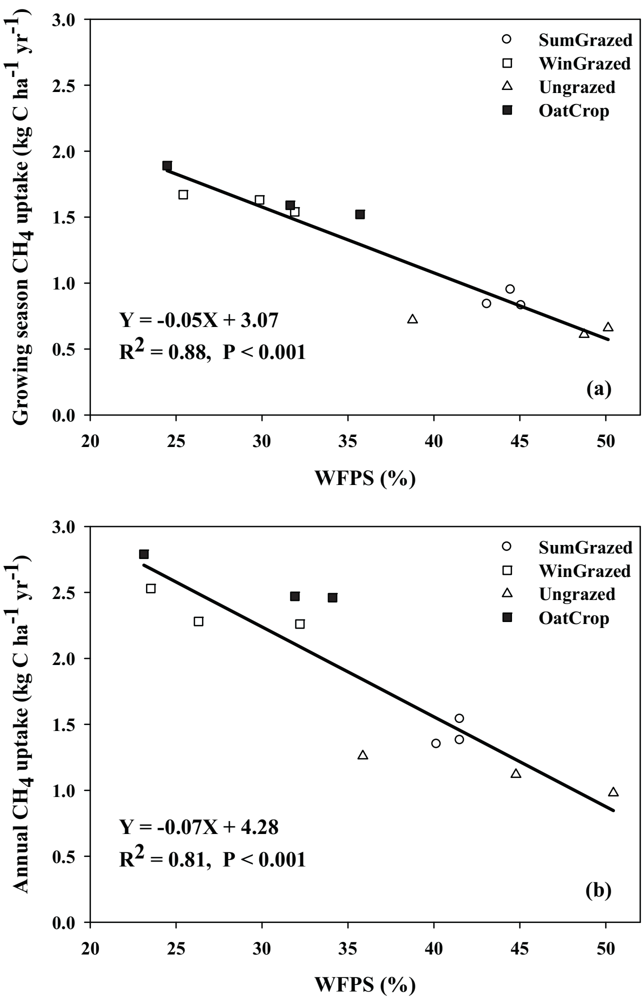

Fig. 9. Regression analysis between cumulative growing season CH_4 uptake (kg C ha^{-1} yr^{-1}) and mean water-filled pore space (%WFPS, 0–6 cm) during the growing season (a), annual CH_4 uptake (kg C ha^{-1} yr^{-1}) and annual mean WFPS (0–6 cm) (b) in all land uses from 30 May 2012 to 30 May 2015.

Ungrazed and OatCrop, respectively, which were within the range of 1.0–2.0 reported from other studies (Butterbach-Bahl and Papen, 2002; Wang et al., 2005; Chen et al., 2010, 2011; Wu et al., 2011). However, they were lower than the Q_{10} values for the non-growing seasons (SumGrazed: 2.3; WinGrazed: 3.1; Ungrazed: 2.4; OatCrop: 3.2), indicating that soil CH_4 consumption in the non-growing season might be more sensitive to future global temperature changes.

Soil moisture controlled CH_4 oxidation through its effect on CH_4 and O_2 diffusion rates in soil and microbial activity of the methanotrophs (Schnell and King, 1996). Both at low and high soil moisture contents, CH_4 uptake capacity may be suppressed, either by physiological water stress of methanotrophs or by restriction of diffusive CH_4 and O_2 transport (van den Pol-van Dasselaar et al., 1998). At all land uses, we observed an inverse correlation between WFPS and daily CH_4 uptake across all three growing seasons (Fig. 7). CH_4 uptake was inhibited by heavy rainfall (Figs. 1–4). Increased soil water content caused by heavy precipitation decreased the diffusion of CH_4 and O_2 into soil and consequently decreased the CH_4 uptake rates. However, the inhibition of CH_4 oxidation by drought stress was absent, since soil moisture in the growing seasons was always greater than the permanent wilting point. Similar negative relationship between soil moisture and CH_4 uptake has been reported in semi-arid grassland in Inner Mongolia (Wang et al., 2005; Liu et al., 2007; Chen et al., 2010, 2011).

5. Conclusions

This study provides three years of measurements of soil-atmosphere CH_4 exchange in a semi-arid agro-pastoral ecotone of northern China. Both of the grasslands and cropland functioned as sinks for atmospheric CH_4 with an average uptake of 1.9 kg C ha^{-1} yr^{-1} (range: 1.0–2.8). The non-growing season contributed 28–43% of annual CH_4 uptake, underlying its importance in estimates of annual CH_4 uptake. Conversion from grassland to cropland and grazing increased CH_4 uptake.

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