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Spatial and temporal patterns of CO_2 and CH_4 fluxes in China's croplands in response to multifactor environmental changes

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ABSTRACT

The spatial and temporal patterns of CO_2 and CH_4 fluxes in China's croplands were investigated and attributed to multifactor environmental changes using the agricultural module of the Dynamic Land Ecosystem Model (DLEM), a highly integrated process-based ecosystem model. During 1980–2005 modelled results indicated that China's croplands acted as a carbon sink with an average carbon sequestration rate of $33.4\,\mathrm{TgC\,yr^{-1}}$ ($1\ \mathrm{Tg} = 10^{12}$ g). Both the highest net CO_2 uptake rate and the largest CH_4 emission rate were found in southeast region of China's croplands. Of primary influences were land-cover and land-use change, atmospheric CO_2 and nitrogen deposition, which accounted for 76%, 42% and 17% of the total carbon sequestration in China's croplands during the study period, respectively. The total carbon losses due to elevated ozone and climate variability/change were equivalent to 27% and 9% of the total carbon sequestration, respectively. Our further analysis indicated that nitrogen fertilizer application accounted for 60% of total national carbon uptake in cropland, whereas changes in paddy field areas mainly determined the variability of CH_4 emissions. Our results suggest that improving air quality by means such as reducing ozone concentration and optimizing agronomic practices can enhance carbon sequestration capacity of China's croplands.

1. Introduction

Anthropogenic emissions of carbon dioxide (CO_2) and methane (CH_4), two important greenhouse gases (GHGs), have the potential to contribute to global warming and ultimately to climate change. It is likely that by 2100, enhanced concentrations of CO_2, CH_4, and other GHGs in the atmosphere will result in an average temperature increase of 1.1–6.4°C (Trenberth et al., 2007). In agricultural ecosystems, the CH_4 emissions have increased by nearly 17% during 1990– 2005 representing about 50% of global CH_4 emissions induced by human activity in 2005 (Clerbaux et al., 2003). As China has 7% of the world's arable land and 21.8% of the world rice area (FAO, 2001), its croplands have the potential to be a major source of CH_4. Therefore, understanding the magnitudes, spatial and temporal patterns of CO_2 and CH_4 fluxes in China's croplands could improve the estimates of the global carbon (C) budget, and provide helpful information that could enhance carbon sequestration capacity in China.

Croplands in China have experienced extensive environmental stress in the past decades due to rapid industrialization, urbanization, climate variability/change including both increasing temperature and extreme weather (Fu and Wen, 1999; Yang et al., 2002), decreased air quality (Akimoto, 2003; Wang et al., 2007), reduced amounts of arable land (from 130 million ha in 1996 to 122 million ha in 2005; Liu et al., 2005) and intensive land management (Li et al., 2005; Lu et al., 2009). Although changes in crop yield, net primary production (NPP), GHGs emissions and C storage that have resulted from these environmental changes have been investigated using inventory, remote sensing and

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models (Chameides et al., 1999; Houghton and Hackler, 2003; Tao et al., 2004, 2008, 2009; Felzer et al., 2005; Li et al., 2005, 2006; Huang and Sun, 2006; Huang et al., 2007; Yang et al., 2007; Zhang et al., 2007b; Yan et al., 2007, 2009b, 2009a; Wang et al., 2008), most previous work considered only one or two environmental factors. Few studies have focused on assessing the response of both CO_2 and CH_4 fluxes in China's croplands to multiple environmental stressors.

Ecological modelling has been proved an effective approach in assessing the effects of multiple environmental factors on terrestrial ecosystems (Tian et al., 2003; Luo et al., 2009). Although many crop models have been developed and applied, most of them (e.g. EPIC and CERES) are site-based, farmer-oriented, lacking of adequate representation of biogeochemical cycles, which have rarely been applied on a large scale (regional or global). Although some biogeochemical models have addressed the effects of multiple factors on biogeochemical cycles on a large scale, they have either generalized all agricultural ecosystems as grasslands or simply ignored managed ecosystems altogether. In addition, other documented models have captured patterns and magnitudes of trace gas emissions (Huang et al., 1998, 2009; Li et al., 2000, 2005, 2006), but few of them have assessed the attribution of multiple factors on large scales. Some recently developed models, for example LPJmL (Bondeau et al., 2007) and Agro-C (Huang et al., 2009), have promoted such work and simulated the carbon (C) budget in agricultural ecosystems at country and global scales. However, little attention has been paid to multifactor-driven changes in C fluxes and pools. This is due in part to a lack of long-term spatial data sets on these environmental factors (including an irrigation and fertilizer application database); in addition, some processes such as ozone (O_3) pollution effect have not been accounted for in these models. For example, O_3 pollution can lead to NPP reduction, weaken the capacity of C sequestration (Ren et al., 2007a) and even result in more carbon loss from agricultural ecosystems with increased fertilizer application (Felzer et al., 2005). Therefore, to reduce uncertainty in estimating the C budget at such a large scale, it is necessary to both develop process-based models that take into account the impacts of major environmental factors on agroecosystems, and to develop relevant regional databases to drive the models.

The Dynamic Land Ecosystem Model (DLEM) is a process-based model that integrated biophysical, biogeochemical, hydrological processes and plant community dynamics, and land management and disturbances into one comprehensive model system, to estimate the fluxes and pools of carbon, nitrogen and water at multiple spatial and temporal scales (Tian et al., 2005, 2008, 2010a,b; Ren et al., 2007a,b; Liu et al., 2008). We improved the DLEM model by integrating a new agricultural module, which addressed the importance of agronomic practices (fertilization, irrigation, rotation, etc.) and climate change on crop growth, phenology and soil biogeochemical cycles in croplands, which provides a tool for studying climate change impact, adaptation and mitigation in agriculture sector. Newly developed, fine resolution, long-term historical datasets including climate, atmospheric CO_2, tropospheric O_3, nitrogen deposition and land-cover and land-use change (LCLUC), were used to drive the model for investigating the patterns and controls of CO_2 and CH_4 in China's croplands during the period 1980–2005. The specific goals of this study were: (1) to quantify the magnitude of CO_2 and CH_4 fluxes in agricultural ecosystems, (2) to analyse spatial and temporal patterns of CO_2 and CH_4 fluxes, (3) to attribute the relative role of environmental factors in the cropland C budget and (4) to identify major uncertainties associated with our estimation of CO_2 and CH_4 fluxes in China's croplands.

2. Materials and methods

2.1. Description of an agricultural module of the DLEM

The DLEM is a highly integrated, process-based terrestrial ecosystem model that aims at simulating the structural and functional dynamics of land ecosystems affected by multiple environmental factors including climate, atmospheric compositions (CO_2, O_3), precipitation chemistry (nitrogen composition), natural disturbance (fire, insect/disease, hurricane, etc.), land-use and land-cover change and land management (harvest, rotation, fertilization, irrigation, etc.). DLEM consists of five vegetation, three soil and seven debris boxes, and couples major biogeochemical cycles, hydrological cycle and vegetation dynamics to make daily, spatially explicit estimates of water, carbon (CO_2, CH_4) and nitrogen fluxes (N_2O) and pool sizes (C and N) in terrestrial ecosystems. DLEM includes five core components: (1) biophysics, (2) plant physiology, (3) soil biogeochemistry, (4) dynamic vegetation and (5) land use and management. This model has been extensively calibrated against various field data covering forest, grassland and cropland from the Chinese Ecological Research Network, US LTER sites and AmeriFlux network (e.g. Ren et al., 2007a,b; Zhang et al., 2007a; Liu et al., 2008; Tian et al., 2010a,b). DLEM has been used to simulate the effects of climate variability and change, atmospheric CO_2, tropospheric O_3, nitrogen deposition and land-cover and land-use change on the pools and fluxes of carbon and water in China (Chen et al., 2006a,b; Ren et al., 2007a,b; Liu et al., 2008), the United States (Zhang et al., 2007a; Tian et al., 2008; 2010a) and the North America (Tian et al., 2010b; Xu et al., 2010).

The DLEM agricultural module enhances the ability of DLEM model to simulate the interactive effects of agronomic practices/land management and other environmental factors on crop growth, phenology and biogeochemical cycles in croplands (Fig. 1). It aims to simulate crop growth and yield, as well as carbon, nitrogen and water cycles in agricultural ecosystems. All the processes of crop growth (e.g. photosynthesis, respiration, allocation) and soil biogeochemistry (e.g. decomposition, nitrification, fermentation) are simulated in the same way as in DLEM for all natural functional types and with a daily time-step.

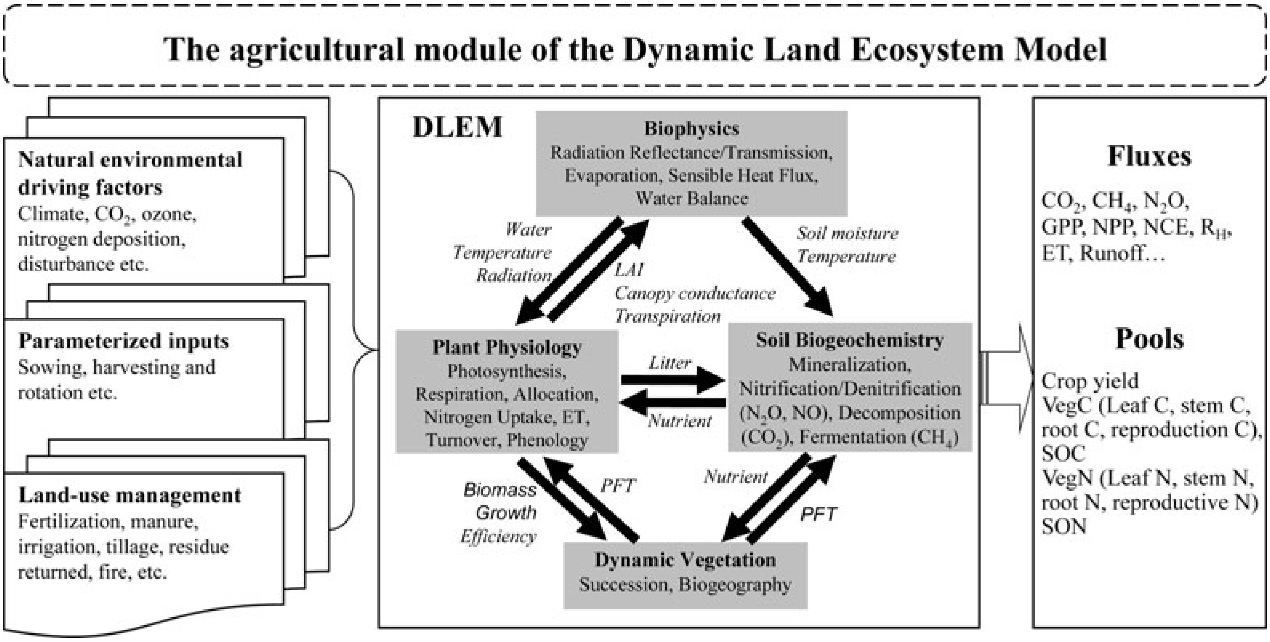


Fig. 1. Framework of the agriculture module of the Dynamic Land Ecosystem Model (DLEM) includes four parts: (1) specific parameterized inputs; (2) process-based biophysics, plant physiology and soil biogeochemistry; (3) natural environmental driving and (4) human management. *Note:* C, N, VegC, SOC and SON represent carbon, nitrogen, vegetation carbon, soil organic carbon and soil organic nitrogen, respectively.

However, in the DLEM agricultural module, different crops are specifically parameterized according to each crop type. Besides natural environmental driving factors, the module gives special attention to the role of agronomic practices, including irrigation, fertilizer application, tillage, genetic improvement and rotation on crop growth and soil biogeochemical cycles. The following provides descriptions of the agricultural module and its applications to China.

2.1.1. *Crops and cropping systems*. We focused on six major crops in China that are representative of both dry farmland and paddy fields or C_3 and C_4 plants, including: corn (irrigated and non-irrigated), rice, wheat (winter and spring), barley and soybean. We used three major cropping systems, including the single cropping system, double cropping system (corn–wheat; rice–rice) and triple cropping system (rice–rice–rice). The main crop categories in each grid were identified according to the global crop geographic distribution map with a spatial resolution of 5 min (Leff et al., 2004), and were then modified with regional agricultural census data derived from FAOSTAT (http://faostat.fao.org/) and the Chinese Academy of Agricultural Sciences (http://www.caas.net.cn). The rotation type in each grid was developed based on phenological characteristics derived from multitemporal remote sensing images with 1 km spatial resolution (Yan et al., 2005), which was then aggregated into 10 km resolution referencing China cropping system census data at the national level and provincial (state) level.

2.1.2. *Phenology*. The phenology information derived from MODIS LAI (with a spatial resolution of 1 km) was used to help identify rotation types. It was calibrated using census data and field data before the application. At the regional level, we simulated crop growth according to the phenological development information, which was developed from a great amount of observations in a number of agricultural meteorological stations and remote sensing-based observations. The role of remote sensing in phenological studies is increasingly regarded as the key to understanding seasonal phenomena in a large area. Phenologic metrics, including start of season, end of season, duration of season and seasonally integrated greenness can be obtained from MODIS time series data and Advanced Very High resolution Radiometer (AVHRR), which has proved useful in determining contemporary patterns at the regional level (e.g. Yu et al., 2005). Our gridded global phenology database developed from MODIS LAI information and field observations was validated against independent field data, and has been used in several studies in the Southern United States, North American continent and Asian regions. In this study, we used substantial observation data from more than 400 of China's agriculture meteorological stations to develop the phenology for each cropping system. Data collection in those stations represented all crop types simulated in this study (Appendix S1). The detailed information about key growth stages were included in the collected database, for example germination, heading, grouting, harvest, etc. which then were used to prescribe the beginning and the end of crop growth stage whereas the dynamic growth rate simulation was limited by daily light, CO_2, temperature, water sources and other environmental factors.

2.1.3. *Agronomic practices*. In this study, the major agronomic practices, including fertilization, irrigation and rotation, were identified and developed using the available data sets. The historical, gridded data set of fertilizer application was developed based on the county-level census data during 1981–2005 and the provincial tabular data during 1950–2005 from National Bureau of Statistics (NBS, http://www.stats.gov.cn/), which was comparable to site observations from Lu et al. (2009). An irrigation map was also developed from the survey database at both county and provincial levels, which changed annually as annual cropland area changed. We assumed that the soil moisture would arrive at field capacity when irrigated, and that the irrigation date is determined when the soil moisture of the top layer drops to 30% of the maximum available water (i.e. field capacity minus wilting point) during the growthing season in the identified irrigated grids. Because the cropping system is very important in China and directly influences estimations of crop production, a contemporary rotation map was developed.

2.1. The carbon budget

In the DLEM agricultural module, the net C exchange (NCE) between agricultural ecosystems and the atmosphere is calculated as

$$\mathrm{NCE} = F_{\mathrm{CO_2}} + F_{\mathrm{CH_4}}, \qquad (1)$$

where $F_{\mathrm{CO_2}}$ is the net C flux related to CO_2, and $F_{\mathrm{CH_4}}$ is the net C flux related to CH_4.

Net ecosystem C flux related to CO_2 is calculated as

$$F_{\mathrm{CO_2}} = \mathrm{GPP} - R_{\mathrm{A}} - R_{\mathrm{H}} - \mathrm{EC} - \mathrm{EP}, \qquad (2)$$

where GPP is gross primary production, a measure of the uptake of atmospheric CO_2 by crops during photosynthesis; R_{A} is autotrophic respiration of crops; R_{H} is heterotrophic respiration associated with decomposition; EC is the C fluxes associated with the conversion of natural areas to agriculture; EP is the decomposition of resulting agricultural and wood products. Net primary production (NPP) is calculated as the difference between GPP and R_{A}. Net ecosystem production (NEP) is calculated as the difference between NPP and R_{H}. The annual NEP of a natural ecosystem is equivalent to its net C storage for the year. Detail functions and processes (related to photosynthesis, respiration and CH_4 simulation) can be found in Appendix S2.

2.2. Other input data

The development of gridded input data sets is an essential component of regional assessment. At minimum, the DLEM agricultural module needs four types of data sets (Table 1, Figs 2 and 3): (1) dynamic crop distribution maps; (2) topography and soil properties (including elevation, slope and aspect; pH, bulk density, depth to bedrock and soil texture represented as the percentage content of loam, sand and silt); (3) climate and atmospheric chemistry (including surface O_3, atmospheric CO_2 and nitrogen deposition) and (4) agronomic practices (including fertilization, irrigation, harvest and crop rotation).

Table 1. Input data used to drive the DLEM agricultural module

Input data	Time	Unit	Description
Climate data	Daily	T: °C d^{-1}; PPT: mm d^{-1}	1980–2005
LCLUC	Annual	0/1 value	1980–2005
Ozone	Daily	AOT40: ppb.h	1980–2005
Nitrogen deposition	Annual	Kg N ha^{-1} yr^{-1}	1980–2005
CO_2	Annual	ppm yr^{-1}	1980–2005
Fertilizer	Annual	gN m^{-2}	1980–2005
Irrigation	Annual		1980–2005
Cropping map	Annual	1–13 value	
Other data set	Soil map, geophysical database, etc.		

Note: T is temperature including maximum, minimum and average temperature; PPT is precipitation; other data set include soil and vegetation information.

Elevation, slope and aspect maps were derived from China's 1 km resolution digital elevation data set (http://www.wdc.cn/wdcdrre). Soil data were derived from the 1:1 million soil maps based on the second national soil survey of China (Shi et al., 2004; Tian et al., 2010c). Daily climate data (including maximum, minimum and average temperature, precipitation and relative humidity) from 1961 to 2000 were developed based on 746 climate stations in China and 29 stations from surrounding countries using an interpolation method similar to that used by Thornton et al. (1997). The atmospheric CO_2 concentration data from 1900 to 2005 were taken directly from Carbon Dioxide Information Analysis Center (Enting et al., 1994; CDIAC, http://cdiac.ornl.gov/). We derived the AOT40 (the accumulated hourly ozone concentrations above a threshold of 40 ppb) data set from the global historical AOT40 datasets constructed by Felzer et al. (2005) and described in detail by Ren et al. (2007a). Nitrogen deposition databases were developed from a regression series by combining the national scale monitoring data, atmospheric transport model results and precipitation distribution in the corresponding year (Lu and Tian, 2007), which has been proved to improve previous work (Denterner et al., 2006) and is comparable to site observations in China (Lu, 2009).

The crop distributional map was derived from the 2000 land use map of China (NLCD2000), which was developed from Landsat Enhanced Thematic Mapper (ETM) imagery (Liu et al., 2005). The potential vegetation distribution map was constructed by replacing the NLCD2000 with potential vegetation from the global potential vegetation maps developed by Ramankutty and Foley (1998). We used long-term land-use history (cropland and urban distribution in China from 1661 to 2000), which was developed by Liu and Tian (2010).

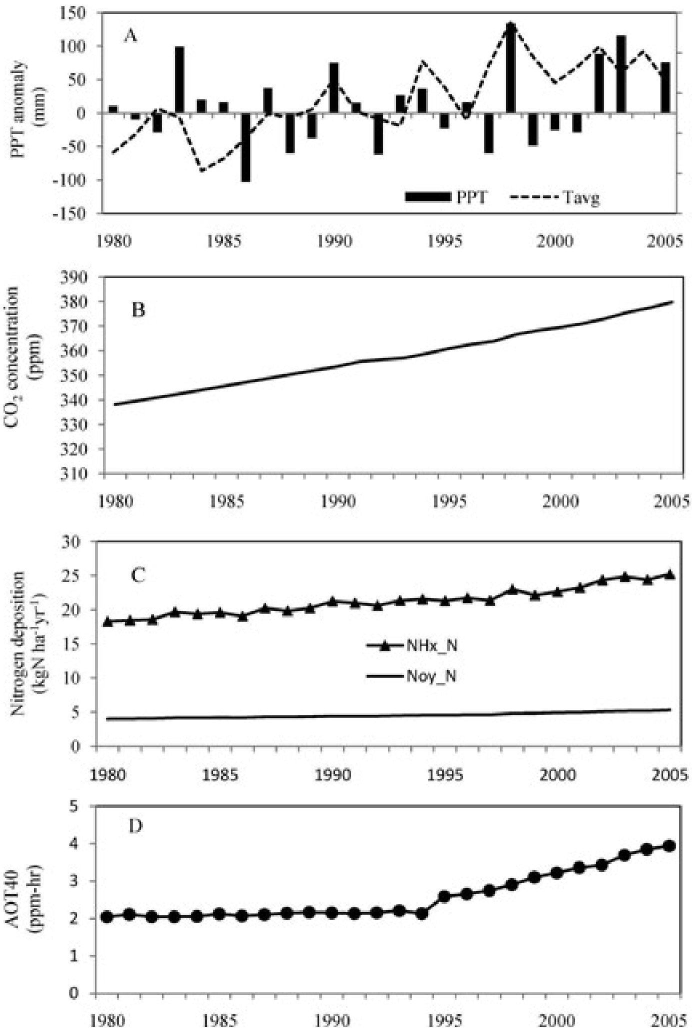


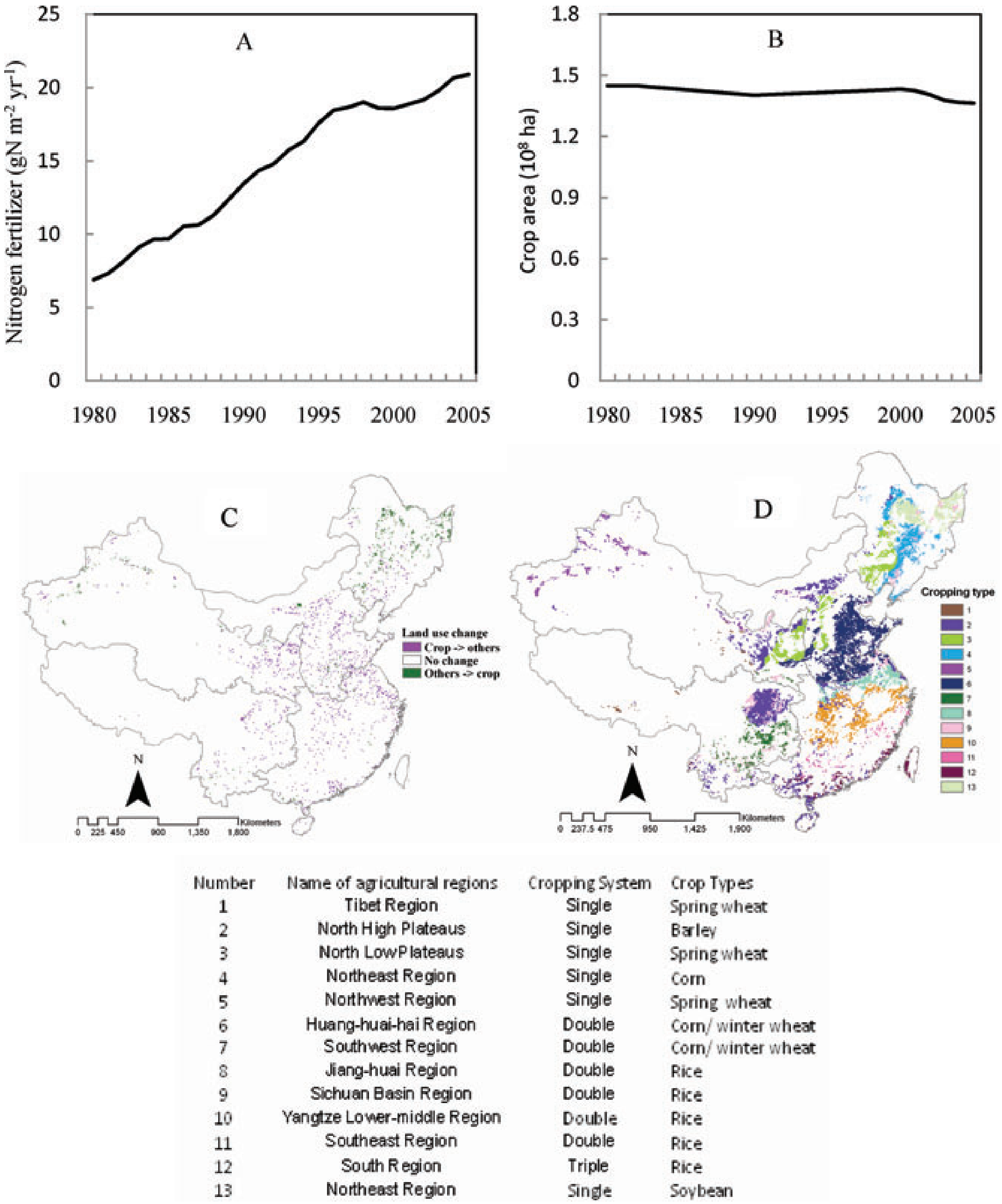
Fig. 2. Multifactor environmental changes in China's agricultural ecosystems. (A) Climate anomalies (precipitation and average temperature relative to 1980–1990 mean data), (B) CO_2, (C) nitrogen deposition and (D) AOT40 (*Note:* AOT40 is a cumulative O_3 index, the accumulated hourly O_3 dose over a threshold of 40 ppb in ppb per hour).

All input data sets have a spatial resolution of 10 km × 10 km; climate and AOT40 data sets were developed on a daily time step whereas CO_2, nitrogen deposition and land-use data sets were developed on a yearly time step.

2.3. Model validation

The simulated results were validated against and compared to independent field data and other studies at both site and regional levels.

2.3.1 Site-level model validation. Two sites, including one dry farmland (rotation of winter wheat and summer maize in Yucheng) and one rice paddy field (two crops of rice in Qingyuan), were selected for model validations (Fig. 4A–F). A comparison of simulated NEP and CH_4 fluxes is shown in Fig. 4A–F. The simulated daily NEP and CH_4 fluxes were comparable to observational data for dry cropland in Yucheng (Fig. 4A–D) and rice paddy in Qingyun (Fig. 4E and F). For NEP of dry cropland in Yucheng, the DLEM agricultural module captured seasonal patterns of daily fluxes, but missed some pulses. Overall, the modelled annual NEP was close to but slightly higher than observed NEP, -827 gC m^{-2} versus -722 gC m^{-2}. Comparisons of modelled CH_4 fluxes and observed CH_4 fluxes in dry cropland (Fig. 4C and D) and rice paddy (Fig. 4E and F) showed DLEM's ability to capture not only seasonal patterns, but also the magnitude of CH_4 fluxes. However, two pulses of CH_4 flux were simulated by the module because of two periods of extremely high precipitation. Further investigation indicated that the first peak of CH_4 emissions was caused by a 2-day heavy rainfall event with a total rainfall of 69.3 mm, and the second peak of CH_4 emissions was associated with a period of heavy precipitation with 60.4 mm day^{-1}. It should be noted that the annual precipitation for Yucheng station was 574 mm in 1997. The DLEM agricultural module also simulated the seasonal pattern of CH_4 fluxes from a rice paddy field in the Qingyun, Southern China. We also compared model results of seasonal CH_4



Number	Name of agricultural regions	Cropping System	Crop Types
1	Tibet Region	Single	Spring wheat
2	North High Plateaus	Single	Barley
3	North Low Plateaus	Single	Spring wheat
4	Northeast Region	Single	Corn
5	Northwest Region	Single	Spring wheat
6	Huang-huai-hai Region	Double	Corn/ winter wheat
7	Southwest Region	Double	Corn/ winter wheat
8	Jiang-huai Region	Double	Rice
9	Sichuan Basin Region	Double	Rice
10	Yangtze Lower-middle Region	Double	Rice
11	Southeast Region	Double	Rice
12	South Region	Triple	Rice
13	Northeast Region	Single	Soybean

Fig. 3. Changes in cropland and nitrogen fertilizer application in China. (A) Annual changes of land management (fertilizer application), (B) cropland area between 1980 and 2005, (C) land-use change in the 1990s and (D) contemporary cropping systems in China.

variations induced by different agronomic practices with field experiments in several sites (Appendix S3).

2.3.2. *Regional comparison with other studies.* The estimation of soil C storage by the DLEM agricultural module was also comparable to other studies (Table 2), although few of these studies were conducted at the national level or for a long historical period. We found that C storage in the soils across China's croplands increased from 1980 to 2005. Our estimations of 16 TgC yr^{-1} for the upper 20 cm across China's croplands and 11.5 TgC yr^{-1} for paddy fields were comparable to Huang and Sun's survey estimation of 18–22 TgC yr^{-1} (Huang and Sun, 2006) and Zhang et al.'s simulation estimation of 4.0–11.0 TgC yr^{-1} (Zhang et al., 2007b) between 1980 and 2000, respectively. The simulated rates of CH_4 emissions at the national level in the 1990s and recent 5 years (2000–2005) were 7.30 and 7.06 TgC yr^{-1}, respectively, which were comparable to other studies whose values ranged from 6.02 to 10.2 TgC yr^{-1} (Bachelet and Neue, 1993; Wu and Ye, 1993; Shen et al., 1995; Huang and Sun, 2006; Wang et al., 2009; Yan et al., 2009b). It is possible that the difference in estimations of CH_4 emissions

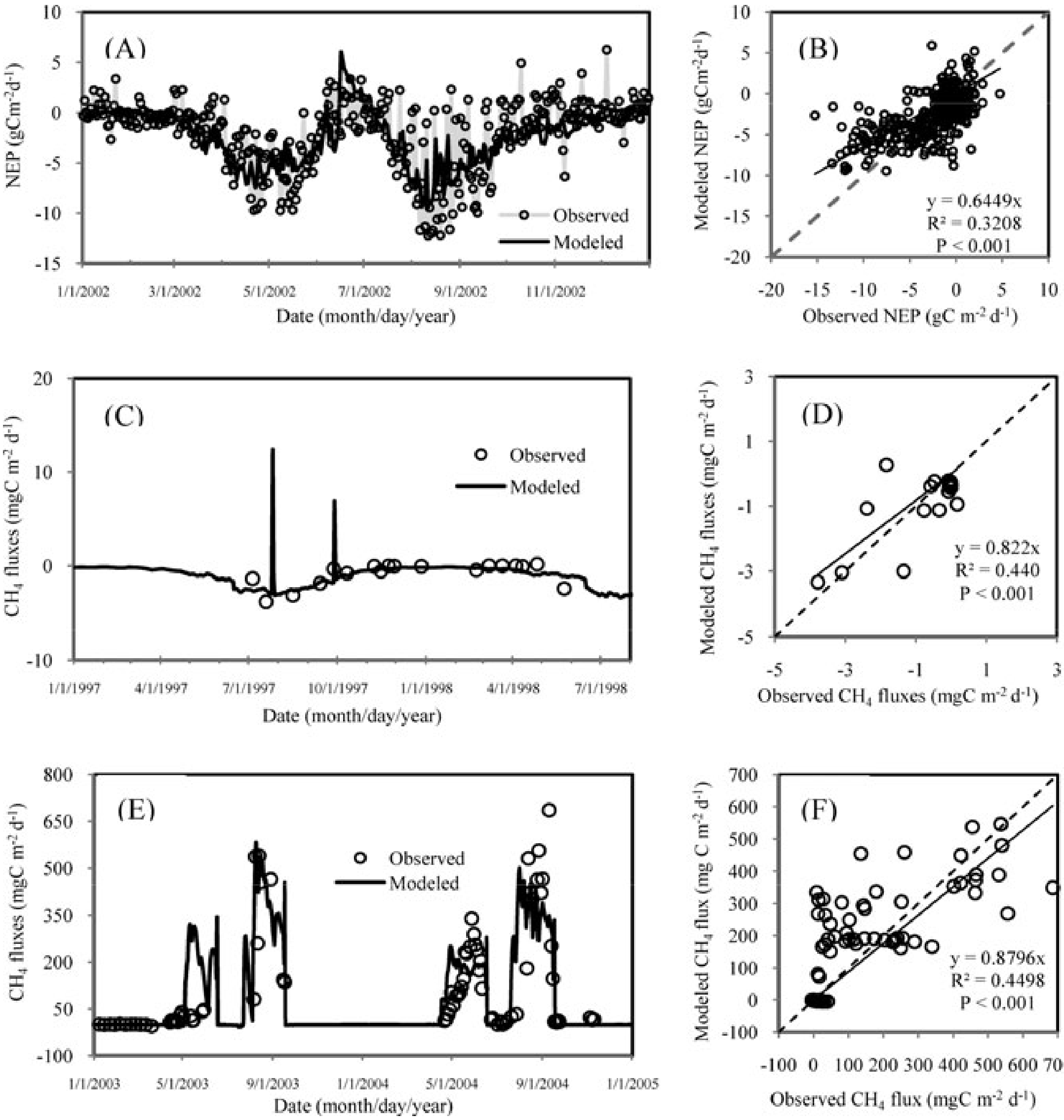


Fig. 4. Comparison of DLEM-estimated CO_2 and CH_4 fluxes with field observations (A: time-series comparison of NEP for dry cropland in Yucheng (36°N, 116°E); B: scattered plot of NEP for dry cropland in Yucheng; C: time-series comparison of CH_4 flux for dry cropland in Yucheng (36°N, 116°E); D: scattered plot of CH_4 flux for dry cropland in Yucheng; E: time-series comparison of CH_4 flux for rice paddy in Qingyuan (23°N, 112°E) and F: scattered plot of CH_4 flux for rice paddy in Qingyuan). Note: the positive indicates release and the negative indicates uptake.

was caused by uncertainty in estimating the paddy area as well as the use of different methods.

2.4. Model simulations

We designed 11 simulations to analyse the relative contribution of each driving factor to the C budget in agricultural ecosystems (Table 3). In simulations one to six, we tried to capture both the direct effects of an environmental factor and the interactive effects of this factor with others on the fluxes of CO_2 and CH_4 in croplands. We conducted five additional simulations (7–11) to test the sensitivity of each factor; in each simulation a particular environmental factor was set to change over time whereas the other environmental factors were held constant at the initial level.

The model was run at a daily time step to simulate crop development and growth. The generic crop parameters, such as phenological development, were developed based on observations from agro-ecosystem experimental stations in China and remote sensing databases. Other parameters, such as light saturated photosynthesis rate and light extinction coefficient, were relatively stable, and could be set for the model default values. Information on agronomic practices (such as sowing, harvest, fertilizer, irrigation and rotation) was obtained from observational and inventory data.

Table 2. Comparisons between modeled and inventory estimations on soil carbon storage in cropland and CH$_4$ emissions from rice paddy in China

Reference	Methodology	Time period	Other study	This study
Huang and Sun (2006)	Inventory	1980–2000	0.018–0.022 PgC yr^{-1} [Increased soil organic carbon (SOC) on national scale on the top soil, 0.2 m]	0.016 Pg Cyr^{-1}
Zhang et al. (2007b)	Modelled	1980–2000	4.0–11.0 TgC yr^{-1} [Increased soil organic carbon (SOC) in rice paddy field]	11.5 TgC yr^{-1}
Wu and Cai (2007)	Inventory	1979–1985	4.4 PgC (national) 1.6 PgC (rice paddy) 2.8 PgC (dry lands) [Soil organic carbon storage (SOC) in top soil layer, 1 m]	4.8 PgC (national) 1.2 PgC (rice paddy) 3.6 PgC (dry lands)

Reference	Area ($\times 10^6$ km^2)	Time period	CH$_4$ emission (TgC yr^{-1})
Yao et al. (1996)	0.32	1991	11.48
Bachelet and Neue (1993)	0.32		$9.11 - 21.62 \times 0.75$
Wu and Ye (1993)			7.02×0.75
Shen et al. (1995)			$10.2 - 12.8 \times 0.75$
Huang et al. (2006)	0.30	2000	6.02
Wang et al. (2009)	0.28	2000–2005	6.25 ± 0.36
Yan et al. (2009a,b)		2000	5.56
This study	0.32–0.37	1990s	7.30 ± 1.08
		2000–2005	7.06 ± 1.15

Table 3. Simulation experiments considering climate, carbon dioxide (CO$_2$), nitrogen deposition (NDEP), ozone (O$_3$), land-cover and land-use change (LCLUC)

	Simulation Experiment	Climate	CO$_2$	O$_3$	NDEP	LCLUC
	Equilibrium	C	C	C	C	C
1	All Combined	H	H	H	H	H
2	All-Climate	C	H	H	H	H
3	All-CO$_2$	H	C	H	H	H
4	All-O$_3$	H	H	C	H	H
5	All-NDEP	H	H	H	C	H
6	All-LCLUC	H	H	H	H	C
7	Climate only	H	C	C	C	C
8	CO$_2$ only	C	H	C	C	C
9	N deposition only	C	C	H	C	C
10	Ozone only	C	C	C	H	C
11	LCLUC only	C	C	C	C	H

Note: H and C stand for historical (H) and constant (C).

For the model run at the regional level, the model simulation began with an equilibrium run to develop the baseline C, N and water pools for each grid with a maximum year of 50 000. A spin-up of about 100 years was then applied if the climate variability/change was included in the simulation experiment, and a spin-up of about 1000 years was used if irrigation was not applied in the simulation experiment. Finally, transient model runs of different experiments were conducted using the daily and/or annual input data.

3. Results

3.1. *Changes in multiple environmental factors in China during 1980–2005*

From 1980 to 2005, annual total precipitation and mean temperature showed substantial interannual and decadal variations. Annual precipitation reached a maximum of 960 mm in 2002, with a minimum of 555 mm in 1986. Mean annual temperature showed an observational increase (Fig. 2A). The atmospheric CO$_2$ concentration steadily increased from 338 ppmv in 1980 to 372 ppmv in 2005 (Fig. 2B).

A dramatic increase in tropospheric O$_3$ levels was observed as well. The simulated mean, monthly accumulated O$_3$ concentration above a threshold of 40 ppbv (AOT40) demonstrates an average increase of 9.5 ppb-hr yr^{-1} over the past 26 years, a trend which has accelerated rapidly since the early 1990s (Fig. 2D). This is possibly due to rapid urbanization in China during this period (Liu et al., 2005). The data set showed seasonal variation of AOT40, with the first peak of O$_3$ concentration occurring in early summer and the second in September. Both peaks appeared approximately at critical times (the growing and harvest seasons) for crops in China. The central-eastern section of North China experienced severe O$_3$ pollution, especially in spring and summer. Thus, O$_3$ pollution may have had significant impacts on crop production.

In 2000, the croplands in China were distributed unevenly with its 141 million hectares (Mha, 10^6) consisting of 35.6 Mha of paddy land and 105.5 Mha of dry farming land (Liu et al., 2005). There were more croplands in the northeastern region of China than in the southeast (SE) and northern regions. Although the total cropland area in China decreased by 8.4 Mha during 1980–2005, with the highest reduction (4.5 Mha) in the SE and then (3.4 Mha) in Mid-North region (MN) China, the cropland area in northeast (NE) China continually increased by 2.6 Mha (Liu and Tian, 2010; Figs 3B and C). As land use and land cover changed, intensively agronomic practices were employed across China's croplands. For example, chemical nitrogen fertilizer (CF-N) application rapidly increased from 6.9 to 20.9 gN m^{-2} during 1980–2000, accounting for 30% of the world's fertilizer use. The highest regional increase in average CF-N application occurred in the MN and SE of China.

In this study, the cropland system was further classified into several major crop types (e.g. wheat, rice, corn, etc.) (Fig. 3D). A simplified crop rotation system was developed based on AVHRR/NDVI data, the cropping system map in China (1:18 000 000) and the observational database in National Agrometeorological Stations. Single cropping areas were large in northwest China and were associated with low temperature, high altitude and drought. Multiple cropping systems were mostly concentrated in southeast China where the climate is warm with a long frost-free period, high precipitation and high population density.

3.2. Interannual and interdecadal variability in NCE, CO_2 and CH_4 fluxes

Our simulation results show that China's agricultural ecosystems acted as a net C sink during 1980–2005 (Table 4, Fig. 5). The C sequestration rates of 46.53, 22.05 and 30.50 TgC yr^{-1} were estimated in the 1980s, 1990s and recent 6 years (2000–2005), respectively. Annual NCE between the atmosphere and agricultural ecosystems varied from year to year (Fig. 5); its lowest value was −31.93 TgC yr^{-1} in 2000.

Carbon dioxide uptake was found across China's croplands with a mean uptake rate of 40.87 TgC yr^{-1} during the study period. Carbon dioxide uptake dominated the C budget in China's croplands (Table 4, Fig. 5), with CO_2 uptake rates of 54.51, 29.36 and 37.31 TgC yr^{-1} in the 1980s, 1990s and 2000–2005, respectively. Accordingly, annual CO_2 flux had a similar pattern to annual NCE, but the magnitude was lower, as annual CH_4 emissions reduced carbon sequestration in croplands (Fig. 5).

Table 4. CO_2 and CH_4 fluxes at national level (TgC yr^{-1})

	NCE	CO_2 flux	CH_4 flux		
	Nation	Nation	Nation	Dry farmland	Paddy field
1980s	46.53	54.51	−7.98	0.11	−8.09
1990s	22.05	29.36	−7.31	0.10	−7.41
00–05	30.50	37.31	−6.82	0.12	−6.94
26-year average	33.42	40.87	−7.45	0.11	−7.56

CH_4 emissions occurred at the national level with an average emission rate of 7.45 TgC yr^{-1}. During the period, rice paddy fields released 7.56 TgC yr^{-1} and dry farmlands absorbed CH_4 at a rate of 0.11 TgC yr^{-1} (Table 4, Fig. 5). The total amount of CH_4 emissions decreased slightly, with average estimates of 8.09, 7.41 and 6.94 TgC yr^{-1} for the 1980s, 1990s and 2000–2005, respectively. In summary, CH_4 emissions were offset by increasing CO_2 uptake since the 1980s, which has led to C sequestration in China's croplands in recent decades.

3.3. Spatial variation in CO_2 and CH_4 fluxes in China

Regional analysis shows large spatial variations in CO_2 flux in five regions (Figs 6 and 7). Mean annual CO_2 flux indicates that CO_2 uptake occurred in four regions including the northwest (NW), MN, southwest (SW) and SE, with average uptake rates of 6.19, 17.53, 6.11 and 21.08 TgC yr^{-1}, respectively, which were 15.5%, 42.9%, 14.9% and 51.7% of the total national CO_2 uptake. On the contrary, the northeast region released CO_2 at a rate of 10.05 TgC yr^{-1} over the 26-year simulation period. Results of the mean annual CO_2 flux indicate that the highest C sequestration rates occurred in the 1980s in three regions (NW, MN and SE) covering more than 60% of China's total cropland area, which is consistent with the national pattern during the same time period.

The simulated results show that all five regions released CH_4 to the atmosphere. The CH_4 emission rates of the NW, MN, SW, WE and SE regions and their percentages relative to total national CH_4 emissions were 0.04 TgC yr^{-1} (0.8%), 0.10 TgC yr^{-1} (2.1%), 0.3 TgC yr^{-1} (4.0%), 0.92 TgC yr^{-1} (12.4%) and 6.01 TgC yr^{-1} (80.7%), respectively. The SE region was the main source of CH_4 emissions and released at 6.54 TgC yr^{-1} from croplands into the atmosphere in the 1980s. This is the highest emission rate among the five regions for the simulation period (Figs 6 and 7). The main reason for the large spatial variability of CH_4 emissions is that CH_4 emissions was mostly concentrated in fields that were unevenly distributed in the five regions.

Our analysis shows that extreme weather conditions such as drought occurred across China (Fig. 8A), which led to large spatial variations in CO_2 and CH_4 fluxes (Fig. 8B and C). The driest year occurred in 1986 (Fig. 2A), with the lowest annual precipitation between 1980 and 2005. The spatial distribution of precipitation anomaly indicated that low precipitation occurred in most areas of the NW and MN regions, but the largest reduction in precipitation (more than 100 mm) occurred in the north plain where dry farmlands were widely distributed (Fig. 8A). Higher precipitation was found in some areas of the SE. The spatial distribution of CO_2 flux anomaly showed that in 1986, CO_2 release occurred in North China where precipitation decreased significantly compared to the 30-year average (Fig. 8B).

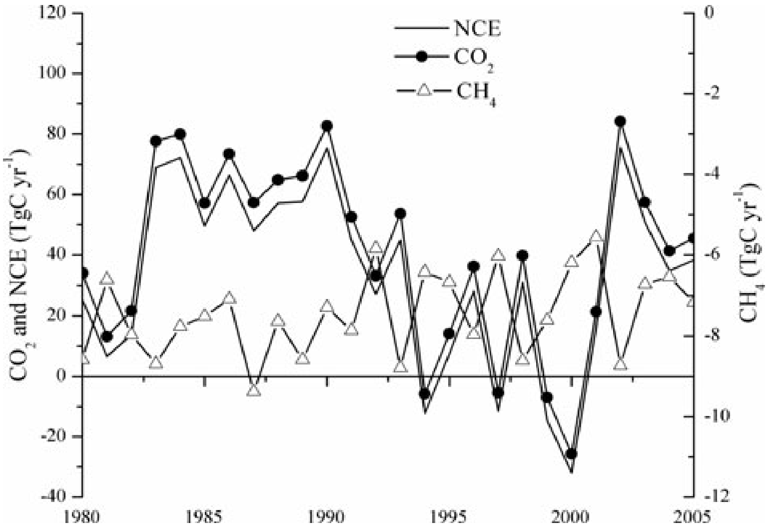


Fig. 5. Annual CO_2 flux, CH_4 flux and net carbon exchange (NCE) during the period 1980–2005 (unit: TgC yr^{-1}). Note: the positive indicates uptake and the negative indicates release.

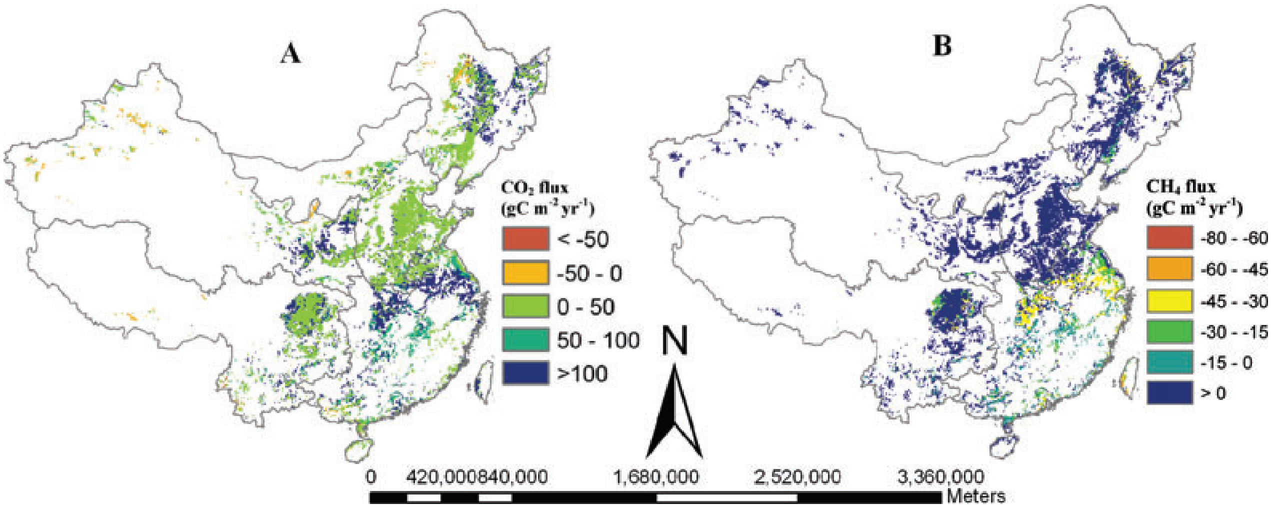


Fig. 6. Spatial distributions of mean annual CO_2 (A) and CH_4 fluxes (B) in recent 5 years (2001–2005). Note: the positive indicates uptake and the negative indicates release.

However, in South China, where precipitation was higher than the 30-year average (1961–1990), we found small changes in CO_2 flux, or even CO_2 uptake. We also found that when irrigation was applied, croplands in the North China plain turned into CO_2 uptake instead of CO_2 release in 1986. Methane emission decreased in 1986 in most of Southern China (Fig. 8C). This was possibly caused by reduced DOC due to an increased soil decomposition rate as a result of additional soil moisture.

3.4. *Relative contributions of multifactor stresses*

We investigated the relative contributions of major environmental factors to the net fluxes of CO_2, CH_4 and NCE in China's agricultural ecosystems. During 1980–2005, the environmental factors controlling CO_2 and CH_4 fluxes changed substantially as described in the section on change in multiple environmental factors in China. Among the five environmental factors affecting NCE between agricultural ecosystems and the atmosphere during the simulation period, our results indicated that LCLUC accounted for 76% of the increase in NCE in the nation over the 26-year period (Fig. 9), with the highest C sequestration rate of 0.04 PgC yr^{-1} in the 1980s. Carbon dioxide and N deposition contributed 43% and 17% of the total NCE increase, respectively. Our simulation results show that both tropospheric O_3 pollution and climate variability/change caused a net C release of approximately 27% and 9% into the atmosphere, respectively. The NCE included net CO_2 flux and net CH_4 flux as shown in eq. (1).

The relative contributions of major environmental factors to CO_2 and CH_4 fluxes varied among regions. The simulated results

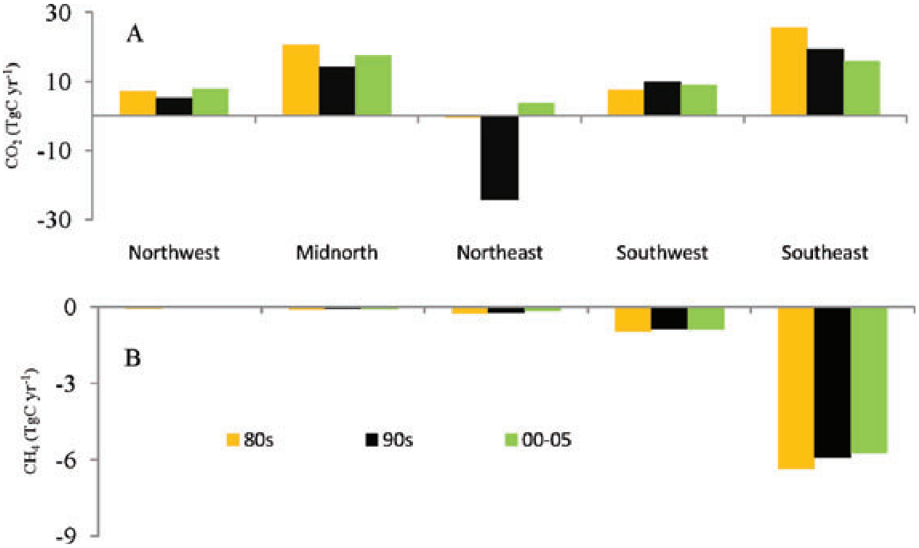


Fig. 7. Mean annual of (A) CO_2 and (B) CH_4 fluxes in different regions during the period 1980–2005. Note: the positive indicates uptake and the negative indicates release.

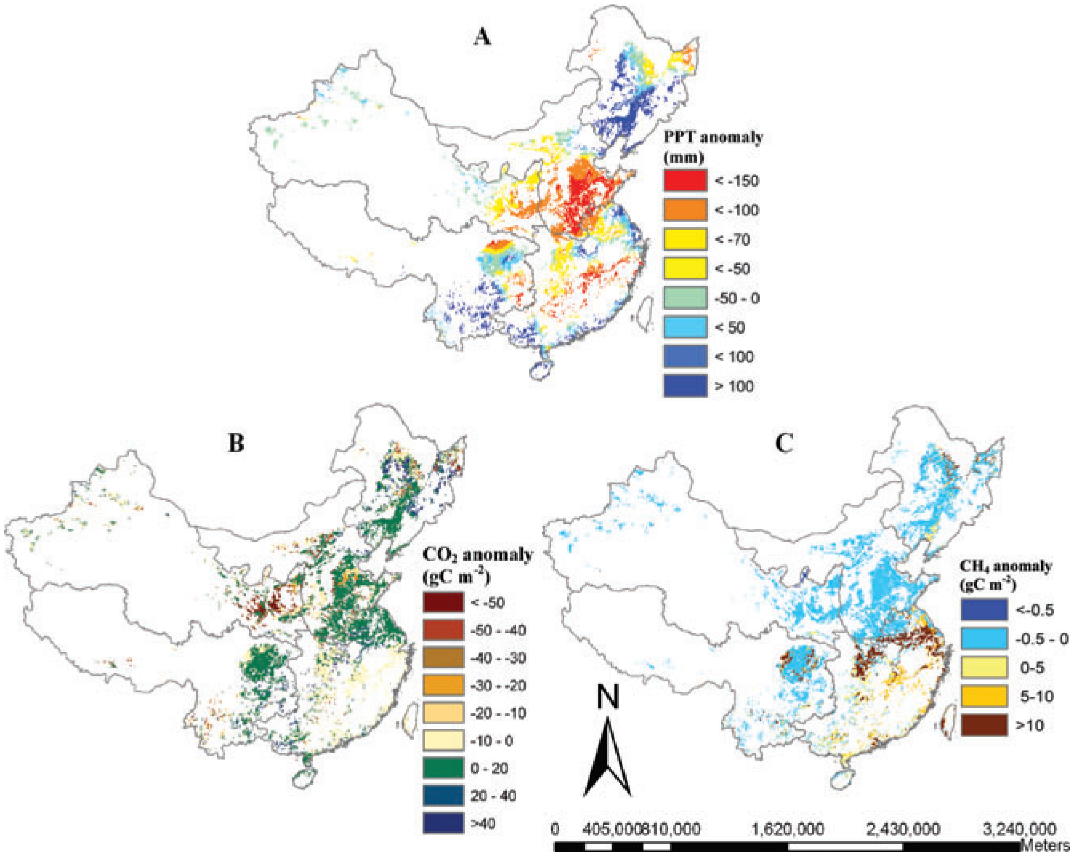


Fig. 8. Spatial distributions of precipitation anomaly (A), CO_2 flux anomaly (B) and CH_4 flux anomaly (C) in extreme dry year 1986. *Note:* anomaly means the difference between precipitation/CO_2 flux/CH_4 flux in 1986 and the 30-year average precipitation/CO_2 flux/CH_4 flux from 1961 to 1990. Note: the positive indicates uptake and the negative indicates release.

Table 5. Decadal mean of CO_2 and CH_4 fluxes (TgC yr^{-1}) in different regions.

	NW	MN	NE	SW	SE
	CO_2/CH_4	CO_2/CH_4	CO_2/CH_4	CO_2/CH_4	CO_2/CH_4
1980s	6.44/−0.07	19.68/−0.12	−0.31/−0.28	3.68/−0.96	27.23/−6.54
1990s	6.12/−0.01	16.35/−0.07	−24.12/−0.23	7.91/−0.90	18.53/−5.88
2000–2005	5.90/−0.01	15.59/−0.10	3.79/−0.15	6.85/−0.89	15.11/−5.51
26-year average	6.19/−0.04	17.53/−0.10	−10.05/−0.30	6.11/−0.92	21.08/−6.01
(%)	(15.5/0.8)	(42.95/2.1)	(−24.6/4.0)	(14.9/12.4)	(51.7/80.7)

Note: NW, MN, NE, SW and SE stand for Northwest, Mid-North, Northeast and Southeast China.

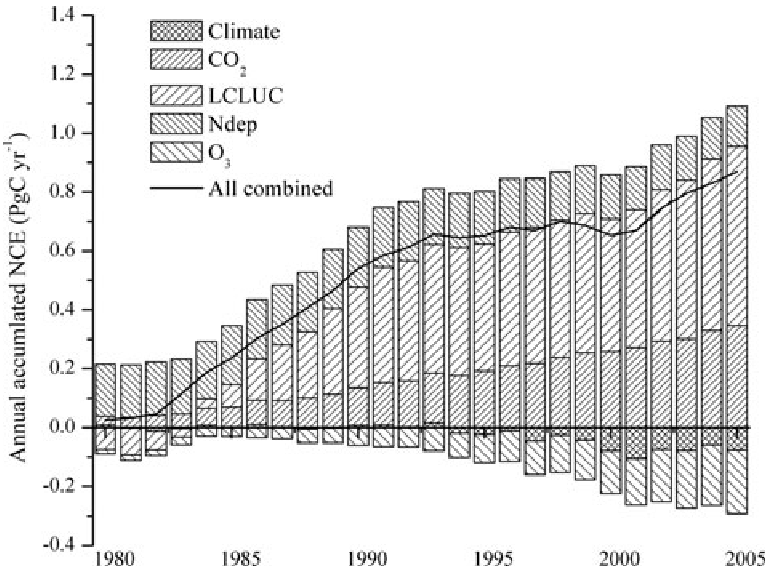


Fig. 9. Annual contribution of climate, CO_2, LCLUC, nitrogen deposition (N_{dep}), O_3 and their combination to accumulated net carbon exchange (NCE) across China's croplands. Note: the positive indicates uptake and the negative indicates release.

showed that the LCLUC had the largest positive effect on CO_2 uptake in all regions except for the NE (Fig. 10). N fertilizer application, a component of LCLUC in this study, increased soil C storage in China's croplands by approximately 0.6 PgC between 1980 and 2005. In the NE region, increase in total soil C storage was primarily due to the expansion of cropland area and the increase in fertilizer application. However, net C released into the atmosphere increased with land conversion from natural ecosystems into agricultural ecosystems, as many other studies have reported (Yang et al., 2007; Ge et al., 2008; Wang et al., 2008). Both elevated atmospheric CO_2 and N deposition caused CO_2 uptake, whereas O_3 pollution and climate variation resulted in CO_2 release in all five regions.

The multifactor effects on CH_4 emissions varied in the five regions (Fig. 10). In the NE region, due to the increase in dry farmlands (Liu et al., 2005), LCLUC resulted in a decrease in CH_4 emissions. In the other four regions, LCLUC increased CH_4 emissions. The largest increase rate was in the SE region as a result of the wide distribution of paddy fields and possibly the abundant DOC derived from C input due to increasing fertilizer application. As discussed earlier, the positive effects of the increasing atmospheric CO_2 which led to C sequestration accelerated CH_4 emissions. The negative effects of O_3 pollution and climate variability/change, which resulted in a C source, restrained CH_4 emissions. Nitrogen deposition stimulated CH_4 emissions in the NE region while resulting in CH_4 uptake from the atmosphere in the other four regions. This was possibly because nitrogen deposition increased crop soil C storage for the expanded cropland area in the NE region. However, increasing N input (N fertilizer application or N deposition) in combination with decreasing cropland area in the other four regions could have sped up soil respiration, or increased CH_4 oxidation, which in turn reduced CH_4 emissions (Xu et al., 2004).

4. Discussion

4.1. *Estimation of CO_2, CH_4 fluxes and net carbon storage*

Globally, the net exchange of CO_2 between the atmosphere and agricultural ecosystems is estimated to be a CO_2 emission of approximately 40 TgC yr^{-1} in the year 2005 (Denman et al., 2007). Using an improved agricultural module of the DLEM, however, we estimated that net flux of CO_2 in China's agricultural

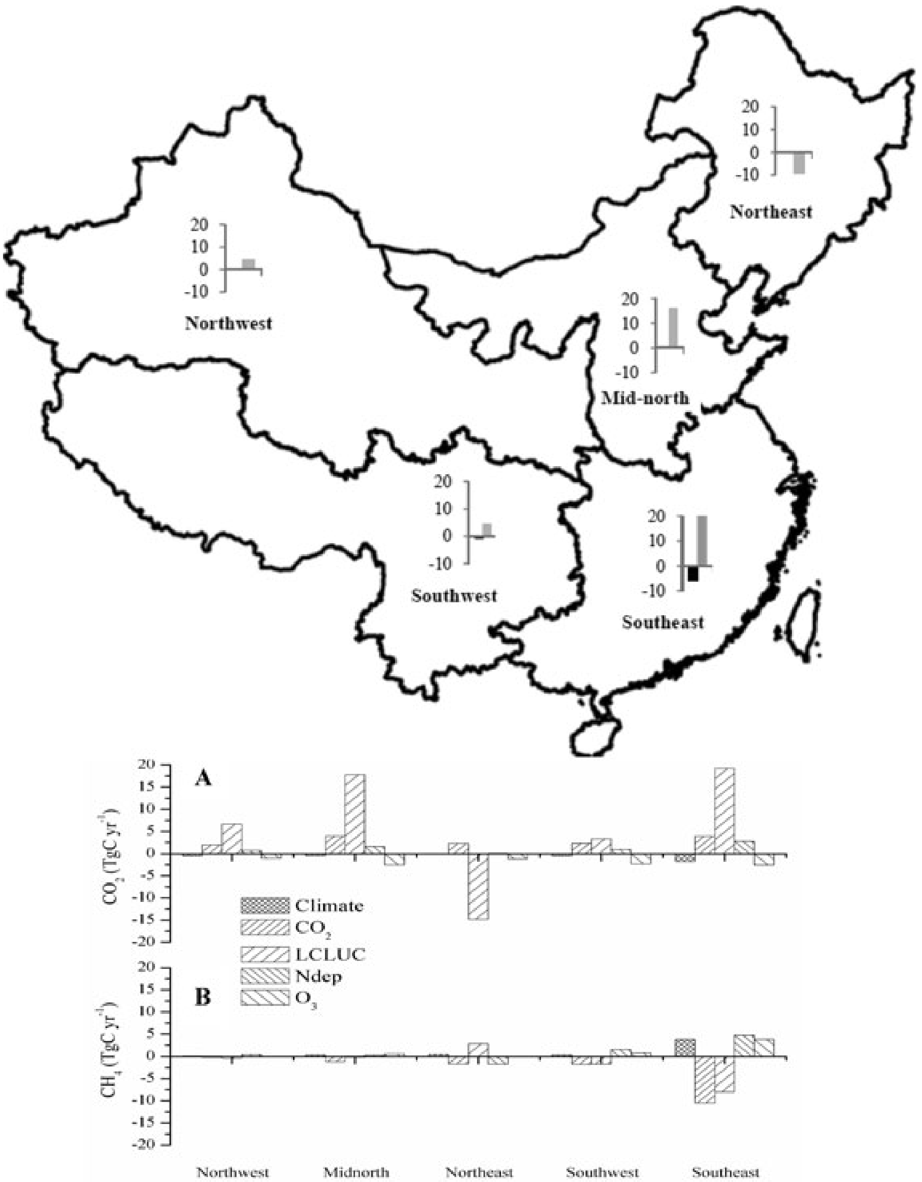


Fig. 10. Mean annual net fluxes of CO_2 and CH_4 (black and grey bars) and changes in mean annual (A) CO_2 and (B) CH_4 fluxes resulted from multiple factors including climate, CO_2, land-cover and land-use change (LCLUC), N deposition (N_{dep}) and O_3 in different regions (texture bars) during the period 1980–2005 (unit: TgC yr^{-1}). Note: the positive indicates uptake and the negative indicates release.

ecosystems was a CO_2 sink of approximately 49.7 TgC yr^{-1} in the year 2005. Possibly, the enhanced CO_2 uptake due to optimized agronomic practices, such as increasing N-fertilizer application, was higher than the CO_2 release that resulted from land conversion and the effects of other environmental factors, such as O_3 pollution. Compared to the global estimation of CH_4 emissions from rice fields of 50 TgC yr^{-1} on average with a range from 31 to 112 TgC yr^{-1} from the 1980s to 1990s (Forster et al., 2007), our estimation of CH_4 release in China's rice paddies of approximately 7.68 TgC yr^{-1} accounted for approximately 15% of the total CH_4 emissions on a global scale, even though China's paddy fields were estimated to be approximately 21.8% of the world rice area (FAO, 2001). Our estimations of both CO_2 and CH_4 fluxes indicate that China's agricultural ecosystems had the high capacity for C sequestration in past decades.

4.2. *Contributions of multiple global change factors to CO_2 and CH_4 fluxes*

Much work has been done on estimating CO_2 and CH_4 fluxes in croplands, but few studies have focused on attributing the effects of the multiple environmental factors which affect these fluxes. Our factorial simulation experiments intended to evaluate the relative contribution of each environmental factor to CO_2 and CH_4 fluxes. Our results indicate that the DLEM agricultural module can capture both the direct effects of an environmental factor and the interactive effects of multiple environmental factors on the net fluxes of CO_2 and CH_4. Below we describe the direct and interactive effects of environmental factors.

4.2.1 Atmospheric components. Increasing CO_2 concentration and N deposition had positive effects on CO_2 uptake, which is consistent with other previous work (e.g. Schindler and Bayley, 1993; Dhakhwa et al., 1997; Tian et al., 1999; Neff et al., 2000). Elevated O_3, in contrast, had a negative effect on C sequestration. This was also consistent with previous work (e.g. Heagle, 1989; Felzer et al., 2005; Sitch et al., 2007). Elevated atmospheric CO_2 resulted in CH_4 emissions from rice paddies due to increasing organic matter; this has been demonstrated in many other studies (Ziska et al., 1998; Allen et al., 2003; Inubushi et al., 2003; Cheng, 2006; Zheng et al., 2006). The simulated results in this study indicate that tropospheric O_3 pollution had negative effects on CH_4 emissions, possibly because it reduced sources of SOM in flooded rice paddy soils due to O_3 negative effects on crop growth (e.g. root exudates and plant debris; Yagi and Minami, 1990; Minoda et al., 1996). Our study showed that N deposition had a positive effect on CO_2 uptake, but its effects on CH_4 emissions were hard to distinguish because it could have had both positive and negative effects on CH_4 emissions (Pancotto et al., 2010).

4.2.2. Climate variability/change. Increasing temperature and changing precipitation during the 26 years of the simulation had complex effects on CO_2 and CH_4 fluxes in China's croplands (Figs 2A, 9 and 10). Simulated fluxes of both CO_2 and CH_4 show substantially interannual variation, which is primarily resulted from climate variability (Vukicevic et al., 2001; Tian et al., 2010b). Variability in temperature and precipitation restricted or stimulated CH_4 emissions depending on its effects on soil decomposition and microbial activity, altered soil moisture, and the quantity and quality of organic matter inputs to the soil (Chapin et al., 2002). The combined effects of temperature and precipitation had a positive impact on CO_2 uptake and negative effects on CH_4 emissions in all five regions. A previous study (Cheng et al., 2008) indicated that increased night temperature reduces the stimulatory effect of elevated CO_2 concentration on methane emission from rice paddy soil. To better address climate impacts on CO_2 and CH_4 fluxes, more factorial simulation experiments are needed to identify the effects of temperature and precipitation, respectively.

4.2.3. The land-cover and land-use management. LCLUC was the dominant factor controlling the temporal and spatial variations of CO_2 and CH_4 fluxes in China's croplands during the study period. Land conversion and fertilizer application were two main components in LCLUC. Previous studies have indicated that LCLUC can play reverse roles in different states; it can lead to C release in the early stages due to land conversion from natural vegetation to cropland (Mann, 1986; Johnson, 1992; Houghton and Goodale, 2004), and then possibly causing C storage increase in soils due to optimized land management such as fertilizer/irrigation application and rotation (Cole et al., 1996) in the late stages when no land conversion has occurred. Our study supports these conclusions as we found that China's croplands acted as a C source due to LCLUC effects in the early 1960s when fertilizer application was small and land conversion occurred in the NE and SW regions. The effects of LCLUC on CH_4 emissions indicate that the reduction of total rice paddy area accounted for a slight decrease in total CH_4 emissions; and the increasing N fertilizer also could reduce CH_4 emissions. Recent study indicates that indirect effects of increased N fertilization on litter quality may reduce final CH_4 emissions (Pancotto et al., 2010). There are still many uncertainties in assessing the net N effect on CH_4 emissions from rice fields on a national or global scale because of this complexity and counter-balancing among the effects. Future study of fertilizer application effects on CH_4 emissions is still needed because N influences every process involved in CH_4 emissions from rice fields, including CH_4 production, oxidation and transport from the soil to the atmosphere, and the interactions among these processes (e.g. Cai et al., 2007; Xie et al., 2009).

Besides, other land management such as water management and residue return to cropland are important factors affecting CO_2 and CH_4 fluxes. Since the 1980s, water management for China's rice paddy fields changed substantially with midseason drainage gradually replacing continuous flooding (e.g. Huke and Huke, 1997). However, water management was simplified in this study because the historical, gridded data (addressing irrigation type, irrigation date and quantities of water use) is still not available for regional scale study. Li et al. (2005) conducted sensitivity analysis on different scenarios of water managements (flooding and mid-season drainage), which indicate that the alterative mid-season drainage method could reduce CH_4 emissions about 40% than traditional flooding method. In our study, we assume that irrigation treatment is conducted in irrigated dry farmland and paddy fields and the irrigation date is identified as the point when the soil moisture of the top layer drops to 30% of the maximum available water (i.e. field capacity minus wilting point) during the growing season. The 'required-irrigation', a kind of optimized water management (mid-season drainage), might cause CH_4 emissions reduction and CO_2 uptake increase, and finally lead to high estimation of carbon sequestration. Also the full popularization of straw return designed in this study could definitely lead to carbon sequestration increase, which was proved by the work of Lu et al. (2009), who reported a reduction of 5.3% in the CO_2 emission due to full popularization of straw return. In addition, tillage practice is an important factor influencing carbon sequestration in China's croplands due to about 80% of total cropland with conventional tillage. However, we did not separate the contribution of tillage practices to total cropland carbon sequestration induced by LCLUC in this study because of a lack of spatial tillage-non-tillage database for driving the model as well as site-specific data for calibrating the tillage-induced soil carbon loss in different cropping systems. Besides, the previous studies have shown controversy results. For example, some studies showed that soil tillage can accelerates organic carbon oxidation releasing high amounts of CO_2 to the atmosphere in a few weeks, especially in the short-term periods (e.g. Ellert and Janzen, 1999; La Scala et al., 2006), however, seasonal and annual monitoring data showed that there was no significant difference in soil CO_2 fluxes between no-tillage and conventional tillage systems (Franzluebbers et al., 1995; Hendrix et al., 1998). Yet, West and Post (2002) pointed out that a change from conventional tillage to no tillage could sequester 14–57 gCm^{-2} yr^{-1} averagely, based on a global database of 67 long-term agricultural experiments with 276 paired treatments. Thus, our estimations of CO_2 and CH_4 emissions from crop soil might be underestimated due to no-tillage strategy application in this study.

4.3. *Uncertainty and future work*

This study has provided the first simultaneous estimation of both CO_2 and CH_4 fluxes in response to historical multiple environmental changes in China's croplands. Although this relatively comprehensive analysis was intended to identify the relative contribution of multifactor controls to CO_2 and CH_4 fluxes in China's croplands, it is also critical to recognize the uncertainties that are inherent in such a study.

Our goal was to expand the range of environmental factors, most of which (e.g. climate, CO_2, land use change) are normally included in model analyses while the rest of them (e.g. O_3 and nitrogen deposition) are new and excluded in previous modelling analyses. But, we should recognize that the mechanisms acting in real ecological systems, especially for intensive managed agroecosystems, are very complicated. For example, other factors such as agronomic practices (e.g. water management, residue return to cropland), natural disturbance (e.g. insect pests), and other air pollution components (e.g. aerosol) also affect carbon sequestration potential in croplands. Although the processes included in the agricultural module of the DLEM model have addressed most important responses, some processes such as responses of carbon allocation and stomatal conductance to elevated O_3 exposure may be important, which have not been represented well in the current DLEM agricultural module. The dynamic response of phenology development to global warming and the relationship between delayed CH_4 production and flooding scenario are also important processes to characterize the temporal patterns of CH_4 fluxes. Nevertheless, the integration of existing information into regional carbon estimation can be an important contribution to scientific understanding. Future information obtained from a network of eddy covariance, multifactorial field experiments, remote sensing observations can be used for model development, application and evaluation. The last but not the least important is the reliability of regional input data, which affects the accuracy of assessing regional C budget with ecosystem models. Because of the complicated cropping systems and land management practices in China, therefore, it is necessary to develop regional input data with higher temporal and spatial resolutions for cropland study in the future.

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Supporting Information

Additional supporting information may be found in the online version of this article:

Appendix S1: Characteristics of field sites in China (selected).
Appendix S2: Description of processes related to photosynthesis, respiration, and methane simulation in DLEM model.
Appendix S3: Comparison of simulated versus observed seasonal patterns of methane emission with different agronomic practices.