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# Attribution of spatial and temporal variations in terrestrial methane flux over North America

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**Abstract.** The attribution of spatial and temporal variations in terrestrial methane ($CH_4$) flux is essential for assessing and mitigating $CH_4$ emission from terrestrial ecosystems. In this study, we used a process-based model, the Dynamic Land Ecosystem Model (DLEM), in conjunction with spatial data of six major environmental factors to attribute the spatial and temporal variations in the terrestrial methane ($CH_4$) flux over North America from 1979 to 2008 to six individual driving factors and their interaction. Over the past three decades, our simulations indicate that global change factors accumulatively contributed $23.51 \pm 9.61$ T g $CH_4$-C ($1$ Tg $= 10^{12}$ g) emission over North America, among which ozone ($O_3$) pollution led to a reduced $CH_4$ emission by $2.30 \pm 0.49$ T g $CH_4$-C. All other factors including climate variability, nitrogen (N) deposition, elevated atmospheric carbon dioxide ($CO_2$), N fertilizer application, and land conversion enhanced terrestrial $CH_4$ emissions by $19.80 \pm 12.42$ T g $CH_4$-C, $0.09 \pm 0.02$ T g $CH_4$-C, $6.80 \pm 0.86$ T g $CH_4$-C, $0.01 \pm 0.001$ T g $CH_4$-C, and $3.95 \pm 0.38$ T g $CH_4$-C, respectively, and interaction between/among these global change factors led to a decline of $CH_4$ emission by $4.84 \pm 7.74$ T g $CH_4$-C. Climate variability and $O_3$ pollution suppressed, while other factors stimulated $CH_4$ emission over the USA; climate variability significantly enhanced, while all the other factors exerted minor effects, positive or negative, on $CH_4$ emission in Canada; Mexico functioned as a sink for atmospheric $CH_4$ with a major contribution from climate change. Climatic variability dominated the inter-annual variations in terrestrial $CH_4$ flux at both continental and country levels. Precipitation played

an important role in the climate-induced changes in terrestrial $CH_4$ flux at both continental and country-levels. The relative importance of each environmental factor in determining the magnitude of $CH_4$ flux showed substantially spatial variation across North America. This factorial attribution of $CH_4$ flux in North America might benefit policy makers who would like to curb climate warming by reducing $CH_4$ emission.

## 1 Introduction

Following carbon dioxide ($CO_2$), methane ($CH_4$) is the second most radiatively important anthropogenic greenhouse gas which contributes approximately 15% (Rodhe, 1990), or even higher (Shindell et al., 2005), to the increases in radiative forcing caused by anthropogenic release of greenhouse gases to the atmosphere (Lelieveld and Crutzen, 1992; Forster et al., 2007). Current regional estimates of $CH_4$ flux, however, are far from certain not only because of the complexity of biotic and abiotic processes responsible for the production and consumption of $CH_4$ (Bousquet et al., 2006; Conrad, 1996), but also because of the limitations and uncertainties in the approaches used for estimations (Denman et al., 2007; Tian et al., 2010a); for example, the uncertainties in the methods of up-scaling and down-scaling (Chen and Prinn, 2006; Liu, 1996), biases in observational data (Sellers et al., 1997; Song et al., 2009; Moosavi et al., 1996), and the uncertainties caused by weakened high spatial heterogeneity of ecosystem properties in the regional estimation of $CH_4$ flux (Frolking and Crill, 1994; Mastepanov et al., 2008; Ding et al., 2004a). Process-based modeling approach has become more and more important in regional estimation of $CH_4$ flux because it bases on the understanding of biogeochemistry of

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CH$_4$ production and consumption, and incorporates the effects of spatial and temporal heterogeneities of major environmental controls on CH$_4$ processes (Tian et al., 2010a; Potter et al., 2006; Potter, 1997; Walter et al., 2001; Zhuang et al., 2004).

One of the most challenging issues for process-based modeling approach, however, is the gap between reality and "virtual reality" in models for simulating all major processes and environmental factors responsible for CH$_4$ production and consumption (Schimel, 2001; Tian et al., 2008; Conrad, 1996). The controlling factors for CH$_4$ production and consumption have been identified as substrates including dissolved organic carbon, CO$_2$, and methanol, and environmental factors including soil pH, oxygen concentration, moisture, temperature, and nitrate concentration etc. (Mer and Roger, 2001; Conrad, 1996). In the globally changing environment, a number of factors may change these substrates and/or environmental factors and further alter CH$_4$ production and consumption; for instance, elevated atmospheric CO$_2$ may enhance CH$_4$ emission by stimulating CH$_4$ production (Hutchin et al., 1995) or reduce CH$_4$ oxidation in soils (Phillips et al., 2001); O$_3$ pollution might suppress CH$_4$ emission (Morsky et al., 2008); climate change may increase or decrease CH$_4$ emission (Cao et al., 1998; Frolking and Crill, 1994; Martikainen et al., 1993); N input (Ding et al., 2004b) including N deposition (Steudler et al., 1989) and N fertilization (Zou et al., 2005) might increase (Börjesson and Nohrstedt, 1998; Bodelier et al., 2000) or decrease (Mer and Roger, 2001; Liu and Greaver, 2009; Steudler et al., 1989) CH$_4$ oxidation; and changes in land cover types may increase or decrease CH$_4$ flux, depending on the direction of land conversion (Willison et al., 1995; Huang et al., 2010; Jiang et al., 2009).

In the changing world to which multiple global change factors contribute individually or in combination (Heimann and Reichstein, 2008), attributing the variations in regional terrestrial CH$_4$ flux to these global change factors is of great significance for understanding atmospheric CH$_4$ dynamics and for policy-making to curb the increase in atmospheric CH$_4$ concentration. Yet, most previous process-based modeling efforts did not simultaneously take into account the effects of these global change factors in the estimations of regional CH$_4$ flux (Cao et al., 1998; Potter, 1997; Zhuang et al., 2007). For instance, Zhuang et al.'s studies only considered the effects of climate variability, rising atmospheric CO$_2$, and land classification; other factors including changes of land cover, N deposition, and O$_3$ pollution, were not considered (Zhuang et al., 2004, 2007); most other studies even simulated solely the effects of climate variability (Cao et al., 1998; Potter, 1997; Walter et al., 2001). Given the complicated effects of multiple global change factors on CH$_4$ production and oxidation (Amaral et al., 1998; Börjesson and Nohrstedt, 1998; Mer and Roger, 2001), and high spatial and temporal heterogeneities of global change factors (Denman et al., 2007; Heimann and Reichstein, 2008), it is urgent to si-

multaneously incorporate multiple global change factors into the simulation of CH$_4$ flux for evaluating the relative contributions from each factor to the spatial and temporal variations in terrestrial CH$_4$ flux at large scale (Bousquet et al., 2006).

North America, one of the extensively studied continents on CH$_4$ budget, is still short of quantification on the relative contributions from global change factors to terrestrial CH$_4$ flux (Bridgham et al., 2006; Potter et al., 2006). In our previous study (Tian et al., 2010a), the continental and country-level fluxes of CH$_4$ over North America's terrestrial ecosystems during 1979–2008 have been estimated by using a process-based ecosystem model, Dynamic Land Ecosystem Model (DLEM), driven by multiple global change factors including climate variability, rising atmospheric CO$_2$, O$_3$ pollution, N deposition, land use change, and N fertilizer application. In this study, we will advance our analysis with emphasis on the attribution of the spatial and temporal variations in terrestrial CH$_4$ flux to multiple global change factors at both continental and country levels.

Specifically, the objectives of this study are (1) to examine the factorial contributions to the spatial variation of terrestrial CH$_4$ flux over North America during 1979–2008; (2) to quantify the factorial contributions to the temporal variations in terrestrial CH$_4$ flux over North America during 1979–2008; (3) to quantify the factorial contributions to the 30-year accumulated fluxes of CH$_4$ over North America at both continental and country levels; and (4) to identify the major factors responsible for the spatial and temporal variations in terrestrial CH$_4$ fluxes at both continental and country levels. The global change factors that will be evaluated in this study include climate variability, elevated atmospheric CO$_2$, N deposition, O$_3$ pollution, changes in land use and land cover types, and N fertilizer application. The interactive effects among these six factors were calculated by subtracting the changes in CH$_4$ flux resulted from the combined effects of changes in CH$_4$ flux caused by individual effect from each factor (see Experiment design section for the detail information).

## 2 Materials and methods

### 2.1 Brief description of the model used in this study

The model used in this study is called the Dynamic Land Ecosystem Model (DLEM) which couples major biogeochemical cycles, hydrological cycles, and vegetation dynamics to make daily, spatially-explicit estimates of carbon (C), nitrogen (N), and water fluxes and pool sizes in terrestrial ecosystems (Tian et al., 2008, 2010a, b; Ren et al., 2007; Liu et al., 2008; Zhang et al., 2007). The DLEM also simulates the managed ecosystems including agricultural ecosystems, plantation forests and pastures. The spatial data set of land management, such as irrigation, fertilizer
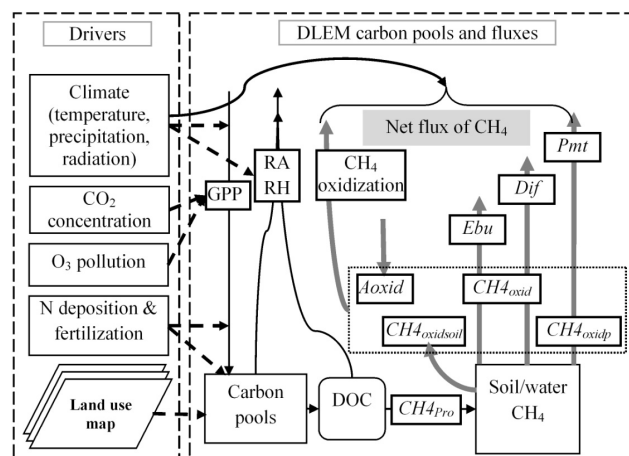
application, rotation, and harvest can be used as input information for simulating influences of land management on the structure and functioning of ecosystems. This model has been calibrated against various field data from the Chinese Ecological Research Network (CERN), US Long-Term Ecological Research (LTER) network, and AmeriFlux network which cover various ecosystems, including forests, grasslands, shrub, tundra, desert, wetland, and croplands. The simulated results have been compared with independent field data and satellite products. The DLEM operates at a daily time step and at a variety of spatial scales ranging from meters to kilometers, from regional to global. The detailed information for DLEM could be referred to our previous publications (Chen et al., 2006; Liu et al., 2008; Ren et al., 2007; Zhang et al., 2007a; Ren, 2009; Zhang, 2008; Lu, 2009; Tian et al., 2010b; Xu, 2010), and the CH$_4$ module has been described in detail in Tian et al. (2010a).

The methane module in the DLEM model mainly simulates the production, consumption, and transport of CH$_4$ (Fig. 1). Due to the relatively small contribution from other substrates (Conrad, 1996; Mer and Roger, 2001), DLEM only considers the CH$_4$ production from dissolved organic carbon (DOC), which is indirectly controlled by environmental factors including soil pH, temperature and soil moisture content. The DOC was produced through three pathways, GPP allocation, and side products from soil organic matter and litter-fall decomposition. CH$_4$ oxidation, including the oxidation during CH$_4$ transport to the atmosphere, CH$_4$ oxidation in the soil/water, and atmospheric CH$_4$ oxidation on the soil surface, is determined by CH$_4$ concentrations in the air or soil/water, as well as soil moisture, pH, and temperature. Most CH$_4$-related biogeochemical reactions in the DLEM were described as the Michaelis-Menten equation with two coefficients: maximum reaction rate and half-saturated coefficient. Three pathways for CH$_4$ transport from soil to the atmosphere including ebullition, diffusion, and plant-mediated transport, are considered in the DLEM (Tian et al., 2010a).

Multiple global change factors yield direct and/or indirect impacts on CH$_4$ processes as simulated in the DLEM (Fig. 1), which could be expressed as the following equation.

$$F_{CH_4} = V_{maxprod} f(C_a, w, T_{air}, APAR) f(O_3) f(N) - V_{maxoxid} f(T_{soil}, WFPS) \quad (1)$$

where $F_{CH_4}$ is the CH$_4$ flux; $V_{maxprod}$ is the maximum rate of CH$_4$ production; $f(C_a, w, T_{air}, APAR)$ describes the indirect effects of atmospheric CO$_2$ concentration, soil moisture, air temperature, and absorbed photosynthetically active radiation on CH$_4$ production through their effects on photosynthesis; $f(O_3)$ describes the indirect effects of O$_3$ pollution on CH$_4$ flux via its effects on photosynthesis; $f(N)$ describes the indirect effects of N input on CH$_4$ production through its impacts on photosynthesis and ecosystem respiration; $C_a$ is atmospheric CO$_2$ concentration, $w$ is soil moisture; $T_{air}$ is



Major processes: Aoxid: Atmospheric CH$_4$ oxidation; CH$_4$ pro: CH$_4$ production; CH$_4$ oxid: CH$_4$ Oxidation during diffusion and ebullition transport; CH$_4$ oxidp: CH$_4$ oxidation during plant-mediated transport; CH$_4$ oxidsoil: CH$_4$ oxidation in soil; Dif: CH$_4$ diffusion transport; Ebu: CH$_4$ ebullition transport; Pmt: Plant-mediated transport of CH$_4$ (Occur in herbaceous wetland only); GPP is the gross primary production, RA is the autotrophic respiration from plant, and RH is the heterotrophic respiration; DOC is the dissolved organic carbon. Drivers are the multiple global change factors which yield controls on or feedback to ecosystem processes in the DLEM framework. The effects from drivers were expressed as the line starting from drivers to ecosystem processes or pools. Solid lines represent direct, while dash lines represent indirect impacts on CH$_4$ processes.

**Fig. 1.** Conceptual diagram showing major processes for CH$_4$ production, oxidation and transport from the soil/water to the atmosphere in response to multiple global change factors.

air temperature, APAR is absorbed photosynthetically active radiation. $V_{maxoxid}$ is the maximum rate of CH$_4$ oxidation, which could be each of three oxidation processes simulated in the DLEM; $f(T_{soil}, WFPS)$ describes the direct effects of soil temperature and moisture on CH$_4$ oxidation; $T_{soil}$ is soil temperature, WFPS is water filled pore space. It should be noted that WFPS is directly related to precipitation. Meanwhile, soil temperature, pH and moisture directly influence CH$_4$ production, while O$_3$ pollution and N input indirectly influence CH$_4$ oxidation through their impacts on ecosystem processes. The impacts of land conversion on CH$_4$ flux could be caused by land-conversion-induced alterations in either substrate or environmental factors. It should be noted that the above equation solely summarizes the direct and indirect effects of multiple global change factors on CH$_4$ processes; some other environmental factors which might influence CH$_4$ processes were not included in this equation, for example, soil pH, soil texture etc.

## 2.2 Study area and input data

North America was selected in this study. It includes United States of America (USA), Canada, and Mexico, covering a total area of approximately 24.71 million km$^2$, about 4.8% of the planet's surface or 16.5% of its land area. Excluding water body and river, the North America consists of 21 237 grids, at a spatial resolution of 32 km × 32 km, which is consistent with North American Regional Reanalysis (NARR) dataset.

We developed gridded, geo-referenced, time-series input data sets of climate (including daily temperature, precipitation, humidity, and solar radiation), annual N deposition rate, annual land-cover change and land management practices (including fertilizer application, irrigation) for the entire continent. The climate dataset was generated based on NARR dataset (Mesinger et al., 2006). NARR data were provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at http://www.esrl.noaa.gov/psd/. The maximum, minimum and average temperatures were calculated based on eight 3-h averages in one day. Precipitation, solar radiation, and relative humidity were directly derived from the NARR dataset. Land-use and land-cover change data were extracted from a global data set, History Database of the Global Environment (HYDE 3.0) (Klein and van Drecht, 2006). O$_3$ pollution data was retrieved from a global dataset developed by Felzer et al. (2005). Annual N deposition data were retrieved from a global data set that was extrapolated from three yearly maps (Dentener et al., 2006). Soil property data, including soil texture, soil pH, soil bulk density, were extracted from a global data set, Global Soil Data Task, which is posted online in the Oak Ridge National Laboratory Distributed Active Archive Center (www.daac.ornl.gov). Fertilizer application data for North America was developed by combining several data sources, including Food and Agriculture Organization (FAO) country-level data (www.fao.org), United State county-level data (www.usda.gov), and Canada provincial-level data (www.cfi.ca). All the datasets were transformed and re-projected to one projection system for driving the DLEM. The annual atmospheric concentration of CO$_2$ before 1959 was estimated by The Vegetation/Ecosystem Modeling and Analysis Project (VEMAP), and the data after 1959 were provided by National Oceanic and Atmospheric Administration (NOAA) (www.esrl.noaa.gov). The spatial distribution of potential vegetation types was developed using different sources of data, including global land-cover derived from Landsat imageries (De Fries et al., 1998), National Land Cover Dataset 2000 (www.usgs.gov), and global database of lakes, reservoirs and wetland (Lehner and Döll, 2004).

Historical data from 1901 to 2008 are prescribed as transient input data sets in this study. The transient input data include: (1) historical daily climate data from 1901 to 2008 including maximum, minimum and average temperatures, relative humidity, solar radiation, and precipitation; the data from 1901 to 1978 were randomly assigned as one year during 1979–2008; (2) historical annual N deposition from 1901 to 2008; (3) historical annual O$_3$ pollution data from 1901 to 2008; (4) historical atmospheric CO$_2$ concentration from 1901 to 2008; (5) historical cropland and urban distribution from 1901 to 2005; the land use since 2005 was assumed to be unchanged due to the shortage of data; and (6) historical N fertilizer application data for cropland for the time period of 1901–2008.

## 2.3 Experimental design

To determine the relative effects of N deposition, O$_3$ pollution, climate variability, elevated atmospheric CO$_2$, land-use change, and N fertilizer application on the terrestrial CH$_4$ flux over North America, we conducted nineteen simulations in this study (Table 1). One overall simulation was set up to simulate the terrestrial CH$_4$ flux over North America by considering the temporal and spatial dynamics of all six global change factors. Six more simulations were set up to simulate the effects of each individual factor on CH$_4$ flux. For example, to determine the effects of climate variability alone, we ran DLEM using the gridded historical daily data for air temperature including maximum, minimum, and average air temperature, relative humidity, solar radiation, and precipitation, but kept all other five global change factors at the level in 1900: the atmospheric CO$_2$ concentration, N deposition, O$_3$ pollution, and N fertilizer application for cropland were kept constant at the level in 1900 and the land cover type in the year of 1900 (potential vegetation map with cropland and urban land in 1900). To determine the effects of CO$_2$ fertilization alone, we ran DLEM using the historical atmospheric CO$_2$ concentrations, but kept all other five global change factors constant: a 30-year average daily climate data was used to represent the constant climatic data and the potential vegetation map with crop and urban land in 1900 was used to represent the constant land cover type, N deposition, O$_3$ pollution, and N fertilizer application data were kept constant in the year of 1900. For each of the above seven simulations, we set up one corresponding simulation which is the same as the previous simulation except the input data in 1979 was used to drive the post-1979 simulations; this design is used to capture the internal dynamics of the system which will serve as baseline.

Five more simulations were set up to separate the contributions from each single climate variable: precipitation, temperature (maximum, average, minimum), solar radiation, and relative humidity. Four simulations were set up to simulate the contribution from each of four climate variables, and one more was set up as baseline to exclude the contribution from system dynamics; i.e. the post-1979 simulations were fed by 1979 climate data (Table 1).

The implementation of DLEM simulation includes the following steps: (1) equilibrium run, (2) spinning-up run and (3) transient run. In this study, we first used potential vegetation map, long-term mean climate during 1979–2008, the concentration levels of N deposition, O$_3$ pollution, atmospheric CO$_2$ in the year of 1900 to drive the model run to an equilibrium state (i.e. the inter-annual variations are <0.1 g m$^{-2}$ for C storage and <0.1 g m$^{-2}$ for N storage). After the system reaches an equilibrium state, the model was run with an addition of cropland and urban areas for another 3000 years for spinning-up purposes. Finally, the model was run in transient mode with daily climate data, annual CO$_2$ concentration and N deposition inputs from 1901 to 2008 to

**Table 1.** Experimental design for this study.

| Simulation | Climate | Nitrogen deposition | CO$_2$ | O$_3$ | Nitrogen fertilizer | Land conversion |
|---|---|---|---|---|---|---|
| 1 | 1900–2008 | 1900–2008 | 1900–2008 | 1900–2008 | 1900–2008 | 1900–2008 |
| 2 | 1900–1979 | 1900–1979 | 1900–1979 | 1900–1979 | 1900–1979 | 1900–1979 |
| 3 | 1900–2008 | 1900 | 1900 | 1900 | 1900 | 1900 |
| 4 | 1900–1979 | 1900 | 1900 | 1900 | 1900 | 1900 |
| 5 | 1900 | 1900–2008 | 1900 | 1900 | 1900 | 1900 |
| 6 | 1900 | 1900–1979 | 1900 | 1900 | 1900 | 1900 |
| 7 | 1900 | 1900 | 1900–2008 | 1900 | 1900 | 1900 |
| 8 | 1900 | 1900 | 1900–1979 | 1900 | 1900 | 1900 |
| 9 | 1900 | 1900 | 1900 | 1900–2008 | 1900 | 1900 |
| 10 | 1900 | 1900 | 1900 | 1900–1979 | 1900 | 1900 |
| 11 | 1900 | 1900 | 1900 | 1900 | 1900–2008 | 1900 |
| 12 | 1900 | 1900 | 1900 | 1900 | 1900–1979 | 1900 |
| 13 | 1900 | 1900 | 1900 | 1900 | 1900 | 1900–2008 |
| 14 | 1900 | 1900 | 1900 | 1900 | 1900 | 1900–1979 |
| | Climate (Precipitation) | Climate (Maximum, average, minimum temperature) | Solar radiation | Relative humidity | Nitrogen deposition, CO$_2$, O$_3$, nitrogen fertilizer, and land conversion | |
| 15 | 1900–2008 | 1900–1979 | 1900–1979 | 1900–1979 | 1900 | |
| 16 | 1900–1979 | 1900–2008 | 1900–1979 | 1900–1979 | 1900 | |
| 17 | 1900–1979 | 1900–1979 | 1900–2008 | 1900–1979 | 1900 | |
| 18 | 1900–1979 | 1900–1979 | 1900–1979 | 1900–2008 | 1900 | |
| 19 | 1900–1979 | 1900–1979 | 1900–1979 | 1900–1979 | 1900 | |

Note: the time period of 1900–2008 indicates that the data for the time period of 1900–2008 was used in the simulation; while the time period of 1900–1979 indicates that the data for the time period of 1900–1979 was used in the simulations, and the simulations after 1979 was fed by the data of 1979.

**Table 2.** Changing rates of driving factors for DLEM simulations.

| Variables | | Changing rates (Mean ± SD) |
|---|---|---|
| Climate | Maximum temperature (°C a$^{-1}$) | 0.04 ± 0.01* |
| | Minimum temperature (°C a$^{-1}$) | 0.03 ± 0.01* |
| | Average temperature (°C a$^{-1}$) | 0.03 ± 0.01* |
| | Precipitation (mm a$^{-1}$) | 0.65 ± 0.65 |
| | Relative humidity (% a$^{-1}$) | −0.01 ± 0.01 |
| | Solar radiation (W m$^{-2}$ a$^{-1}$) | 0.17 ± 0.03* |
| Others | O$_3$ pollution (ppm-hr a$^{-1}$) | 0.93 ± 0.09* |
| | N deposition (mg m$^{-2}$ a$^{-1}$) | 1.98 ± 0.12* |
| | N fertilizer application (mg m$^{-2}$ a$^{-1}$) | 0.06 ± 0.01* |
| | Atmospheric CO$_2$ concentration (ppm a$^{-1}$) | 1.66 ± 0.02* |

* Indicates the changing rate is significantly different from zero; positive values represent increase through the study period, and negative values represent decrease through the study period.

simulate CH$_4$ flux. Only the outputs between 1979 and 2008 were analyzed to show the spatial and temporal patterns of CH$_4$ flux in North America's terrestrial ecosystems. Urban was treated as grassland, which is the same as in the other terrestrial biosphere models (McGuire et al., 2001). Baseline flux was defined as the CH$_4$ flux during 1979–2008 simulated by DLEM driven by the input data of 1979; the changes thereafter comparing to baseline flux was assumed solely caused by global change factors, individually or in combinations.

### 2.4 Model parameterization and validation

The model parameterization and validation at both site and regional levels have been conducted in our previous study (Tian et al., 2010a); the same parameter sets were used in this study. We will not describe them in detail here.

### 2.5 Statistical analysis

The regression analysis was used in this study to find the long-term changing rates of input data and CH$_4$ fluxes generated by various simulations. All the statistical analyses were conducted by using the software SAS 9.2 and SPSS 17.0 for Windows XP.

**Table 3.** Land area of the major biomes in North America.

| Plant functional type | Tundra | Forest | Shrub | Grassland | Wetland | Desert and others | Cropland |
|---|---|---|---|---|---|---|---|
| Area (million km$^2$) | 4.05 | 6.93 $\sim$ 6.99 | 3.57 $\sim$ 3.59 | 2.61 $\sim$ 2.64 | 2.06 $\sim$ 2.07 | 0.53 $\sim$ 0.60 | 2.51 $\sim$ 2.59 |
| Percentage | 18.09 | 31.10 | 15.98 | 11.72 | 9.23 | 2.49 | 11.39 |

Note: biome-level areas may not sum to totals because of the effects of rounding in reporting those values.

## 3 Results

### 3.1 Spatial and temporal patterns of driving forces during 1979–2008

Regression analysis was performed to estimate the temporal patterns of major input variables during 1979–2008 (Tables 2, 3). For the climatic variables, maximum, minimum, and average temperatures, and solar radiation showed significantly increasing rates of $0.04 \pm 0.01\,°C\,a^{-1}$, $0.03 \pm 0.01\,°C\,a^{-1}$, $0.03 \pm 0.01\,°C\,a^{-1}$, and $0.17 \pm 0.03\,W\,m^{-2}\,a^{-1}$, respectively; yet precipitation and relative humidity did not show any significant change along the study period. All the other driving factors significantly increased since 1979; the long-term increasing rates were $0.93 \pm 0.09\,ppm\text{-}hr\,a^{-1}$ for O$_3$ pollution, $1.98 \pm 0.12\,mg\,m^{-2}\,a^{-1}$ for N deposition, $0.06 \pm 0.01\,g\,m^{-2}\,a^{-1}$ for N fertilizer application, and $1.66 \pm 0.02\,ppm\,a^{-1}$ for atmospheric CO$_2$ concentration, respectively. The area of different land cover types changed slightly through the study period; for instance, the cropland area increased from 2.51 million km$^2$ to 2.59 million km$^2$; the areas of forest, shrub, grassland and wetland changed in very small magnitude. It should be noted that all above statistic were continental-level values; the changes in specific area or specific time period might be quite different.

Spatial variations of input data including potential vegetation distribution, N deposition, N fertilizer application rate, and O$_3$ pollution were shown in Fig. 2. The Fig. 2a shows the contemporary spatial distribution of vegetation used in this study; it is noteworthy that natural wetlands primarily distribute in Alaska, western Canada, south to the Hudson Bay, eastern coastal area, and Florida in the USA (Fig. 2a). The severely O$_3$-polluted area over North America locates in western part of North America such as the northwestern USA which could be as high as more than 5000 ppb-hr (monthly accumulated hourly O$_3$ dose over a threshold of 40 ppb in ppb-hr), while the other areas, especially northern end of continental North America, feature low O$_3$ pollution (Fig. 2b). The major cropland with high N fertilizer application (larger than $10\,g\,N\,m^{-2}\,a^{-1}$) locates in USA, including western, central, and eastern costal area of USA. Canada and Mexico had small amount of cropland and received lower application rate of N fertilizer (Fig. 2c). The high N deposition primarily occurred in eastern part of the continental North America, including southeastern Canada, eastern USA and portions of Mexico (higher than $1\,g\,N\,m^{-2}\,a^{-1}$); while northern Canada features quite low N deposition (lower than $0.01\,g\,N\,m^{-2}\,a^{-1}$) (Fig. 2d).

### 3.2 Spatial distribution of CH$_4$ flux during 1979–2008

The CH$_4$ flux over the entire continent of North America showed substantial spatial variations (Fig. 3); the terrestrial ecosystems acted either as a source of atmospheric CH$_4$ as high as more than $30\,g\,C\,m^{-2}\,a^{-1}$, or as a sink of atmospheric CH$_4$ as high as $1\,g\,C\,m^{-2}\,a^{-1}$. A major source for atmospheric CH$_4$ was found in northwestern part of North America, including southern part of Canada, western part of Canada, north central USA, southeastern USA, and Alaska; a strong sink for atmospheric CH$_4$ was found in the southern part of the continental North America, including southern USA and most of Mexico; and other areas acted as a weak sink of atmospheric CH$_4$.

### 3.3 Factorial contributions to the spatial variation in terrestrial CH$_4$ flux during 1979–2008

In this study, we intended to examine the global change factor-induced changes in CH$_4$ emission since 1979, so we assumed that the annual CH$_4$ emission over North America during 1979–2008 with no driving forces changed is the baseline emission, and the changes in CH$_4$ flux compared to the baseline flux are caused by individual and/or interactive effects of these global change factors. To quantify the factorial contributions to the spatial variations in terrestrial CH$_4$ flux during 1979–2008, we first calculated the global change factor-induced CH$_4$ flux by subtracting annual flux by the baseline flux, and then summed them up to reach the global change factor-induced CH$_4$ flux over 30 years.

Over the past 30 years, climate variability enhanced CH$_4$ emission in northwestern part of North America including western parts of Canada and northwestern USA, while decreased CH$_4$ emission in northern, central, and southern parts of North America (Fig. 4a); N deposition enhanced CH$_4$ emission across large area of North America, primarily in eastern parts of Canada, and southeastern US (Fig. 4b); elevated atmospheric CO$_2$ enhanced CH$_4$ emission over large area of continental North America yet did not yield significant impacts on southwestern US and majority of Mexico (Fig. 4c); O$_3$ pollution exerted no significant effects on CH$_4$ flux across majority of North America, while decreased
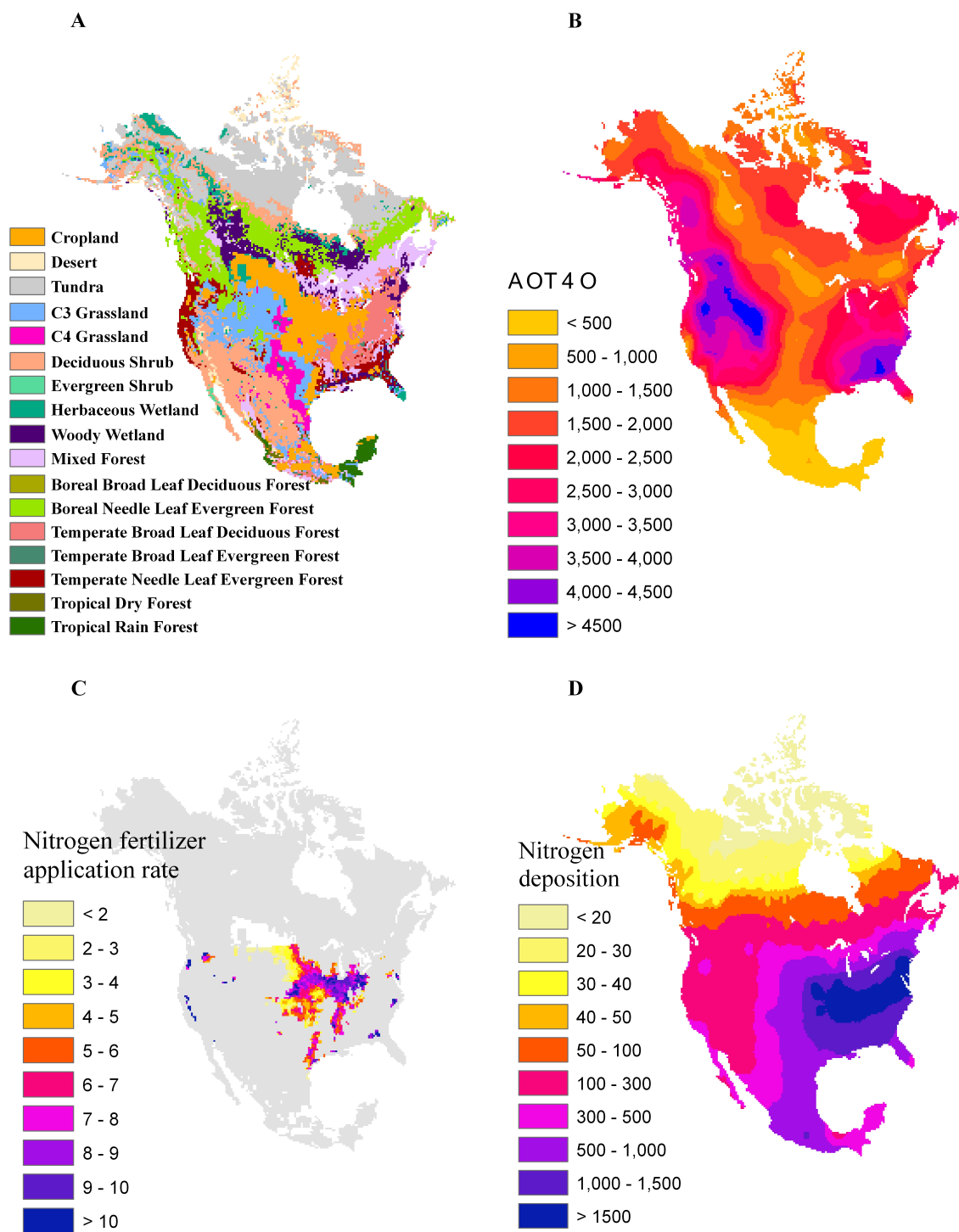
**Fig. 2. (A)** Contemporary vegetation map, and spatial distribution of 30-year averages of **(B)** monthly O$_3$ pollution (ppb–hr), **(C)** N fertilizer application (g N m$^{-2}$ a$^{-1}$), and **(D)** N deposition rate (mg N m$^{-2}$ a$^{-1}$).
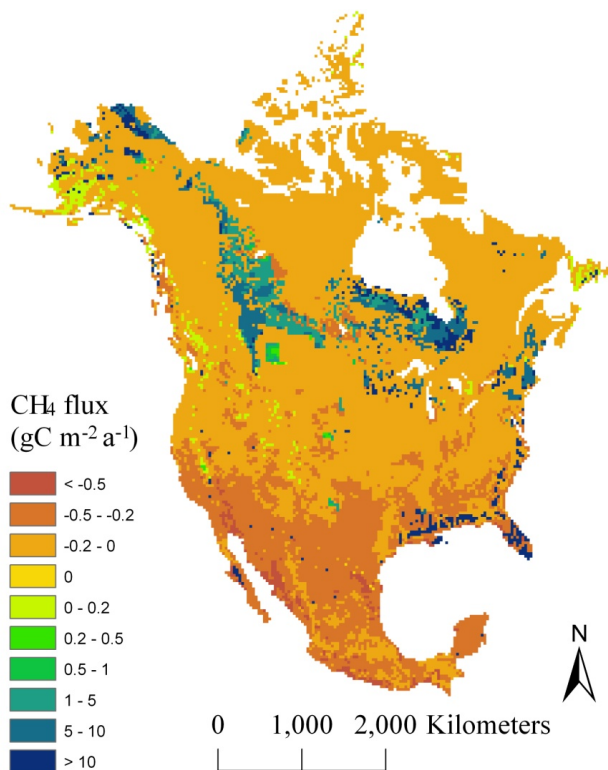
**Fig. 3.** Spatial variations of terrestrial $CH_4$ fluxes caused by global change factors over North America from 1979 to 2008.

$CH_4$ emission in southeastern parts of the continental North America and enhanced $CH_4$ emission in small magnitude over portions of Canada (Fig. 4d); N fertilizer application and land conversion slightly enhanced $CH_4$ emission in portions of agricultural land throughout North America (Fig. 4e and f); interactive effects between/among global change factors enhanced $CH_4$ emission in large area of North America, especially the Southwest (Fig. 4h); combining all the effects from various global change factors, the $CH_4$ emission over the western North America was enhanced over the past three decades, while portions of northern and central North America experienced the reduced $CH_4$ emission (Fig. 4g).

## 3.4 Temporal patterns of $CH_4$ flux during 1979–2008

The $CH_4$ flux over the entire continental North America showed significant inter-annual fluctuations during 1979–2008 (Fig. 5). The lowest annual $CH_4$ emission was $11.74\,T\,g\,CH_4$-$C\,a^{-1}$ in 1998, and the highest was $18.42\,T\,g\,CH_4$-$C\,a^{-1}$ in 2005. Before 2001, the $CH_4$ flux did not show any significant change; however, since 2002 the $CH_4$ emission rate increased dramatically, reached its peak in 2005, and decreased slightly since then. The mean annual $CH_4$ flux over the past 30 years in North America's terrestrial ecosystems was $14.69\pm1.64\,T\,g\,CH_4$-$C\,a^{-1}$; and the overall increasing rate of $CH_4$ flux was $0.10\pm0.03\,T\,g\,CH_4$-$C\,a^{-1}$ over study period ($P=0.003$) (Fig. 5a).

Various global change factors yielded significantly different effects on the long-term trends of continental $CH_4$ flux during 1979–2008 (Fig. 5). Climate variability generated a substantially inter-annual variation in $CH_4$ flux, with an increasing rate of $0.15\pm0.04\,T\,g\,CH_4$-$C\,a^{-1}$ ($P=0.002$) (Fig. 5b). The continuously rising atmospheric $CO_2$ concentration kept accelerating $CH_4$ emission at an overall increasing rate of $0.02\pm0.004\,T\,g\,CH_4$-$C\,a^{-1}$ ($P<0.001$) (Fig. 5d), while $O_3$ pollution decreased $CH_4$ emission at a rate of $0.01\pm0.001\,T\,g\,CH_4$-$C\,a^{-1}$ ($P<0.001$) (Fig. 5e). N deposition generated an increasing rate of $0.71\pm0.05\,Gg\,CH_4$-$C\,a^{-1}$ ($1\,Gg=10^9\,g$) to continental-level $CH_4$ emission ($P<0.001$) (Fig. 5c), while N fertilizer application alone did not exert any significant effects on $CH_4$ flux at the continental scale (Fig. 5f). Land conversion increased the terrestrial $CH_4$ emission over North America from 1979 to 1995, and then decreased it from 1996 to 2008. Over the entire study period, a significantly increasing rate of $0.007\pm0.001\,T\,g\,CH_4$-$C\,a^{-1}$ ($P<0.001$) was simulated for the terrestrial $CH_4$ emission over North America in response to land conversion only (Fig. 5g). A statistically significant correlation was also found between climate-induced annual $CH_4$ flux and the overall $CH_4$ flux contributed from all factors during 1979–2008 ($P<0.001$).

## 3.5 Factorial contributions to the accumulated $CH_4$ flux during 1979–2008 at continental and country levels

To quantify the relative contributions from multiple global change factors to the $CH_4$ flux over North America during 1979–2008, we summed up the individual global change factor-induced changes in $CH_4$ flux over 30 years to analyze the contributions of six single factors and their interaction. To express the uncertainties associated with the accumulated $CH_4$ flux caused by six individual factors and their interaction, we treated the thirty annual fluxes as a sample to calculate the average flux and its standard error. Finally, the 30-year accumulated flux and its standard error over study period were reported. Through the 30-year study period, the accumulated continental $CH_4$ flux over North America was $440.75\pm8.97\,T\,g\,CH_4$-C, of which $417.24\pm6.83\,T\,g\,CH_4$-C was contributed from baseline flux and $23.51\pm9.61\,T\,g\,CH_4$-C was caused by global change factors (Table 4). $O_3$ pollution and the interactive effects between/among multiple factors decreased $CH_4$ emission by $2.30\pm0.49\,T\,g\,CH_4$-C and $4.84\pm7.74\,T\,g\,CH_4$-C, respectively, while all the other single factors increased $CH_4$ emission from North America's terrestrial ecosystems (Fig. 6).

The 30-year accumulated $CH_4$ emission was $214.89\pm3.19\,T\,g\,CH_4$-C for USA and $230.47\pm8.72\,T\,g\,CH_4$-C for Canada, respectively. Mexico acted as a sink for atmospheric $CH_4$, and the accumulative sink strength was $4.62\pm0.19\,T\,g\,CH_4$-C over the past 30 years (Table 4). For USA, climate variability and $O_3$ pollution accumulatively decreased $CH_4$ emission by $3.49\pm9.33\,T\,g\,CH_4$-C and $2.06\pm$
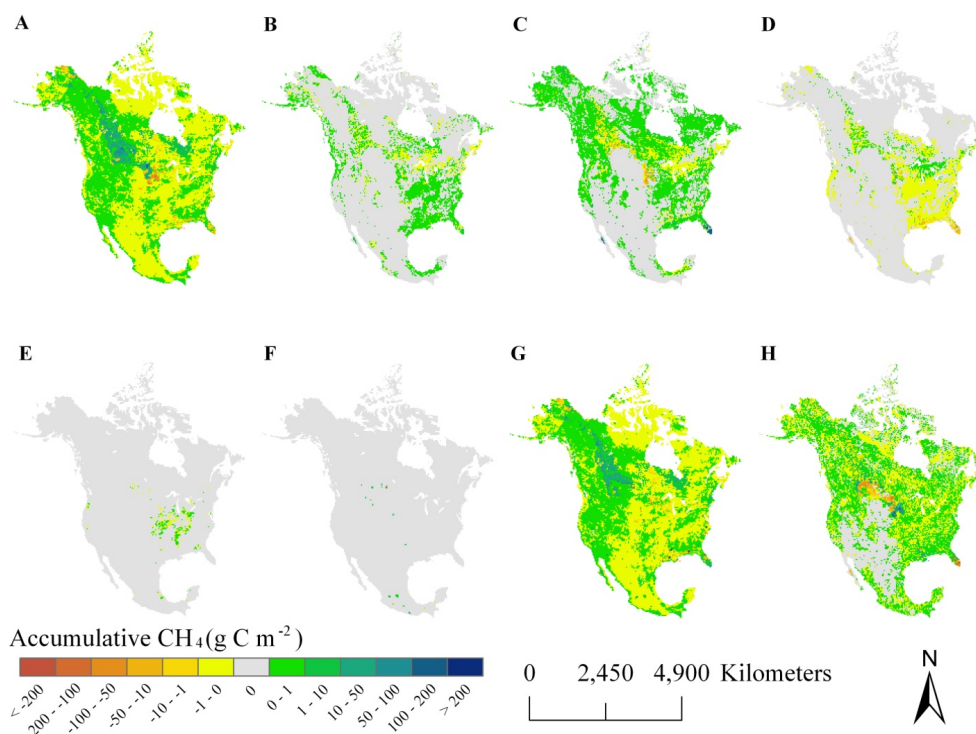
**Fig. 4.** Factorial contributions to the spatial variations in accumulated CH$_4$ flux over North America from 1979 to 2008 ((**A**): climatic variability; (**B**): N deposition; (**C**): CO$_2$; (**D**): O$_3$ pollution; (**E**): N fertilizer application; (**F**): land conversion; (**G**): all combined; (**H**): interaction).
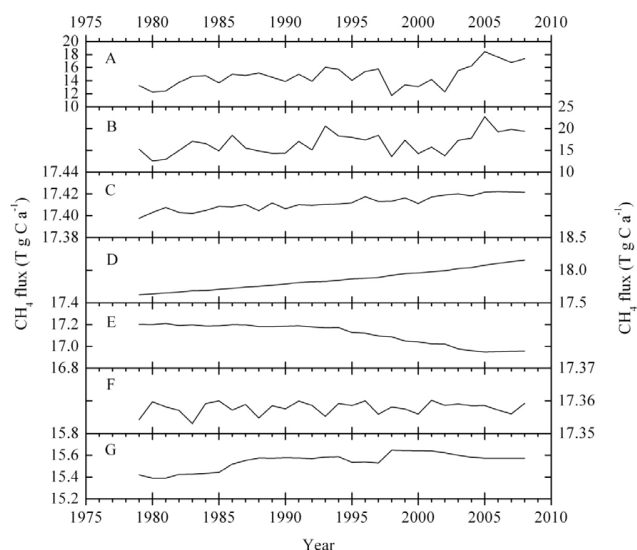


**Fig. 5.** Temporal variations of terrestrial CH$_4$ flux caused by global change factors over North America from 1979 to 2008 ((**A**): all combined simulation; (**B**): climate only simulation; (**C**): N deposition only simulation; (**D**): CO$_2$ only simulation; (**E**): O$_3$ only simulation; (**F**): N fertilizer application simulation; (**G**): land conversion only simulation).

0.44 T g CH$_4$-C, respectively, during 1979–2008, while N deposition, elevated atmospheric CO$_2$ and N fertilizer application, and land conversion accumulatively enhanced CH$_4$ emissions (Table 4). For Canada, it is estimated that climate variability accumulatively enhanced CH$_4$ emission by $23.32 \pm 10.95$ T g CH$_4$-C during 1979–2008, N deposition, O$_3$ pollution, and N fertilizer application increased CH$_4$ emission; while elevated atmospheric CO$_2$, land conversion and multiple-factor interaction decreased CH$_4$ emission (Table 4). All factors except elevated atmospheric CO$_2$ are important for CH$_4$ emission in Mexico; simulation results showed that the elevated atmospheric CO$_2$ accumulatively decreased CH$_4$ consumption by $1.74 \pm 0.20$ T g CH$_4$-C in Mexico during 1979–2008 (Table 4). Precipitation made positive impacts on CH$_4$ flux at continental and country-levels. Relative humidity, solar radiation, temperature, and their interactions also exerted influences, positive or negative on CH$_4$ flux (Table 5). Overall, the global change factors enhanced CH$_4$ emission from USA and Canada, while decreased CH$_4$ uptake from Mexico from 1979 to 2008 (Fig. 7).

For the continental and country-level accumulated CH$_4$ fluxes over 30 years, the baseline emission made the biggest contribution; it accounted for 94.67% of the continental CH$_4$ emission, and 97.78%, 92.34%, and 123.61% of the CH$_4$ fluxes in the USA, Canada, and Mexico (Table 4).

**Table 4.** Factorial contributions to the accumulated CH$_4$ from 1979 to 2008.

|  |  | Baseline | Climate | Ndep | CO$_2$ | O$_3$ | Nfer | Land conversion | Interaction | Total flux |
|---|---|---|---|---|---|---|---|---|---|---|
| US | Accumulated CH$_4$ flux (T g C) | 210.13 ± 2.41 | −3.49 ± 9.33 | 0.05 ± 0.01 | 5.59 ± 0.69 | −2.06 ± 0.44 | 0.004 ± 0.001 | 5.36 ± 0.50 | −0.69 ± 6.90 | 214.89 ± 3.19 |
|  | Percentage (%) | 97.78 | −1.62 | 0.02 | 2.60 | −0.96 | 0.002 | 2.49 | −0.32 | 100 |
| Canada | Accumulated CH$_4$ flux (T g C) | 212.82 ± 6.72 | 23.32 ± 10.95 | 0.001 ± 0.003 | 0.53 ± 0.04 | −0.01 ± 0.01 | 0.001 ± 0.0002 | −1.48 ± 0.14 | −4.71 ± 2.28 | 230.47 ± 8.72 |
|  | Percentage (%) | 92.34 | 10.12 | 0.0004 | 0.23 | −0.003 | 0.0004 | −0.64 | −2.04 | 100 |
| Mexico | Accumulated CH$_4$ flux (T g C) | −5.71 ± 0.21 | 0.003 ± 0.30 | 0.04 ± 0.01 | 1.74 ± 0.20 | −0.24 ± 0.05 | 0.005 ± 0.0003 | 0.08 ± 0.01 | −0.53 ± 0.13 | −4.62 ± 0.19 |
|  | Percentage (%) | 123.61 | −0.07 | −0.80 | −37.64 | 5.19 | −0.10 | 1.64 | 11.44 | 100 |
| North America | Accumulated CH$_4$ flux (T g C) | 417.24 ± 6.83 | 19.80 ± 12.42 | 0.09 ± 0.02 | 6.80 ± 0.86 | −2.30 ± 0.49 | 0.01 ± 0.001 | 3.95 ± 0.38 | −4.84 ± 7.74 | 440.75 ± 8.97 |
|  | Percentage (%) | 94.67 | 4.49 | 0.02 | 1.54 | −0.52 | 0.002 | 0.90 | −1.10 | 100 |

Note: country- or individual factor-based estimates may not sum to totals because of the effects of rounding in reporting those estimates.
Combined represents the effects with all six factors being considered; the Baseline represents contribution from baseline emission; the Climate represents the impacts of climate variability only; Ndep represents the impacts of N deposition; CO$_2$ represents the impacts of CO$_2$ variation; O$_3$ represents the impacts of O$_3$ pollution; Nfer represents the impacts of N fertilizer application; Land conversion represents the impacts of land cover change only; Interaction represents the balance of all interactive effects of the six environmental factors; the positive values represent CH$_4$ emission, while negative values represent CH$_4$ uptake by terrestrial ecosystems.
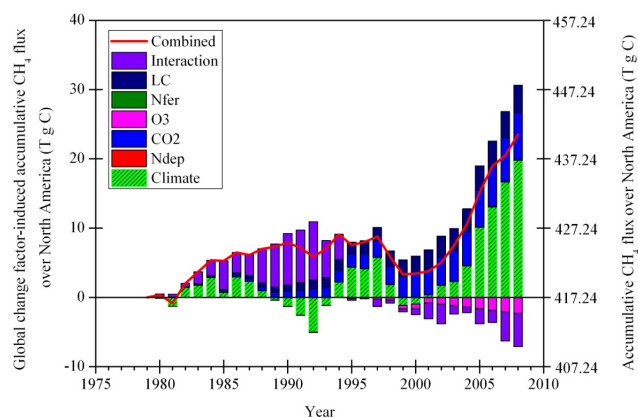


**Fig. 6.** Factorial contributions to accumulated CH$_4$ flux over North America during 1979–2008 (The right Y-axis shows the accumulated CH$_4$ flux with baseline; Interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; Nfer means contribution from N fertilizer application; O$_3$ means contribution from O$_3$ pollution; CO$_2$ means contribution from elevated atmospheric CO$_2$; Ndep means contribution from N deposition; Climate means contribution from climate variability).

## 3.6 Factorial contributions to the inter-annual variations in CH$_4$ flux during 1979–2008 at continental and country levels

Inter-annual variation is one of major attributes of ecosystem processes; it may be caused by internal mechanisms or external environmental controls. Inter-annual variation in terrestrial CH$_4$ has been shown over North America during 1979–2008 (Fig. 5). After removing the baseline emission of CH$_4$, we identified the major factors for the year-by-year variation in CH$_4$ flux (Fig. 8). Over the study period, climate variability and multiple-factor interaction played a predominant role

in contributing to the inter-annual fluctuation in terrestrial CH$_4$ flux (Fig. 8). Climate variability-induced effects dominated the increases in CH$_4$ emission over four time periods: 1981–1984, 1993–1995, 1998–1999, and 2004–2008. Over the time period of 1987–1990, the interaction among multiple global change factors dominated the sink of atmospheric CH$_4$. During other time periods, multiple-factor interaction also made significant contributions to the changes in CH$_4$ flux although it did not dominate the inter-annual fluctuations in CH$_4$ flux. Of the climate impacts on inter-annual variations in terrestrial CH$_4$ fluxes, we further conducted multiple linear regressions to partition the contributions from each climate variable. All variables including precipitation, relative humidity, solar radiation, and temperature made significant contribution, with the largest contribution from precipitation.

After partitioning continental flux into country-level fluxes of CH$_4$, we further analyzed and identified the major factors controlling the inter-annual fluctuations in terrestrial CH$_4$ flux over each country. It is found that the major factors leading to inter-annual fluctuation in terrestrial CH$_4$ flux varied across countries. Climate variability and multiple-factors interaction dominated the inter-annual fluctuations in terrestrial CH$_4$ flux in USA; for instance, the climate variability dominated the sink of atmospheric CH$_4$ over USA during the periods of 1988–1995; multiple-factor interaction dominated the sink of atmospheric CH$_4$ over USA in the year of 2007 (Fig. 9a). Climate variability outweighed other factors in controlling the increases in terrestrial CH$_4$ emission over Canada (Fig. 9b). Climate variability and interactive effect of multiple-factor affected the inter-annual fluctuations in terrestrial CH$_4$ flux over Mexico; since 1996, although the elevated atmospheric CO$_2$ outweighed other factors in contributing to the decrease in terrestrial CH$_4$ consumption, climatic variability dominated the inter-annual fluctuation in CH$_4$ flux over Mexico (Fig. 9c). Further analysis showed that all climate variables made significant contributions, with

**Table 5.** Contributions from individual climate variable to the climate-induced CH$_4$ accumulation (T g C) from 1979 to 2008.

| | Precipitation | Relative humidity | Solar radiation | Temperature | Interaction | Total flux |
|---|---|---|---|---|---|---|
| US | 14.99±8.26 | −5.30±6.71 | 2.57±6.76 | 0.55±6.66 | −16.30±20.54 | −3.49±9.33 |
| Canada | 8.53±8.71 | −28.02±6.95 | −32.54±7.15 | −18.48±6.91 | 93.82±21.90 | 23.32±10.95 |
| Mexico | 0.49±0.23 | 0.81±0.22 | 0.59±0.21 | −0.08±0.28 | −1.81±0.63 | 0.003±0.30 |
| North America | 24.01±10.08 | −32.51±9.01 | −29.38±9.45 | −18.01±9.00 | 75.69±29.14 | 19.8±12.42 |

Note: country- or individual factor-based estimates may not sum to totals because of the effects of rounding in reporting those estimates.

the largest contribution from precipitation in the USA, and Canada, and temperature in Mexico.

## 4 Discussion

### 4.1 Comparisons with others

Over the study period of 1979–2008, continental North America experienced significant environmental change (Wofsy and Harriss, 2002), which was also reflected in the input data for simulations in this study (Figs. 2, 3). These significant changes in environmental factors altered the regimes of terrestrial CH$_4$ flux over North America at both temporal and spatial scales. Spatial heterogeneity in terrestrial CH$_4$ flux is primarily determined by land use type over North America. The relatively high CH$_4$ emission in northwestern continental North America is due to the dense distribution of natural wetland in that region (Fig. 2a) (Bridgham et al., 2006); the strong CH$_4$ sink in the south part of continental North America is due to the tropical forests and high air temperature which are usually associated with high CH$_4$ oxidation rate (Amaral et al., 1998; Curry, 2009; Ridgwell et al., 1999). The strong sources of CH$_4$ in northeastern and southeastern US are consistent with Potter et al.'s study (Potter et al., 2006).

We also compared our model results against previous studies to verify our simulated factorial effects on CH$_4$ flux for major biomes (Table 6). DLEM-derived continental-average response to elevated CO$_2$ is a 58% increase in CH$_4$ emission for wetland, which is close to the middle point of a previously reported range of $0 \sim 146\%$, and is a 1% decrease in CH$_4$ consumption for meadow grassland, which is comparable to Kanerva et al. (2007) result that shows a negative yet not significant effect of elevated atmospheric CO$_2$ on CH$_4$ consumption in a meadow ecosystem. Model-estimated results show that elevated atmospheric CO$_2$ decreased CH$_4$ consumption in temperate forest at a rate of 3%, which is lower than $9 \sim 30\%$ as reported from previous field studies (Phillips et al., 2001; Ambus and Robertson, 1999); this is probably due to one or several of three reasons: the scarcity of data in previous studies, preference to report unusual value in field experiments, and the different methods used in this

research and other studies. The effects of O$_3$ pollution on CH$_4$ flux were comparable between our continental estimations and previous studies; both agreed that the O$_3$ pollution exerted negative yet not significant effects on CH$_4$ from peatland and meadow grassland (Table 6).

DLEM-derived N input effects on CH$_4$ emission or uptake are quite consistent with previously summarized results in dry cropland. Model-estimated N deposition-induced CH$_4$ emission is $7.43 \pm 1.09\,\mathrm{mg\,C\,m^{-2}\,a^{-1}}$ per $\mathrm{g\,N^{-1}\,m^{-2}\,a^{-1}}$ for dry cropland, comparing to $12 \pm 6\,\mathrm{mg\,C\,m^{-2}\,a^{-1}}$ per $\mathrm{g\,N^{-1}\,m^{-2}\,a^{-1}}$ summarized in Liu and Greaver's study (2009). However, it is fairly different between DELM-estimated and summarized N input effects on CH$_4$ flux for other biomes. For example, model-estimated N deposition-induced CH$_4$ uptake is $-0.32 \pm 0.02\,\mathrm{mg\,C\,m^{-2}\,a^{-1}}$ per $\mathrm{g\,N^{-1}\,m^{-2}\,a^{-1}}$ for forest, compared to $17 \pm 5\,\mathrm{mg\,C\,m^{-2}\,a^{-1}}$ per $\mathrm{g\,N^{-1}\,m^{-2}\,a^{-1}}$ in Liu and Greaver's study (2009), and $-10.75 \pm 3.98$ ($\mathrm{mg\,C\,m^{-2}\,a^{-1}}$ per $\mathrm{g\,N^{-1}\,m^{-2}\,a^{-1}}$) in CH$_4$ uptake in a field experiment (Steudler et al., 1989). It should be noted that the changes in CH$_4$ flux result from net changes in CH$_4$ production and consumption; for example the increases in CH$_4$ emission might result from either increases in CH$_4$ production or decreases in CH$_4$ consumption; the increases in CH$_4$ uptake might result from either increases in CH$_4$ oxidation or decreases in CH$_4$ production; Liu and Greaver's study solely reported production or uptake (2009), while this study reported the net flux from production, oxidation, and transport (Materials and methods section).

The differences in model-estimated and summarized N effects on CH$_4$ flux in forests might be due to a few reasons: the missing mechanisms in our model, lacking of field observations in summarization, or the different methods in two studies. N restrain on methanotrophy, long been identified as one of the most important mechanisms for the effects of N impact on CH$_4$ flux (Dunfield and Knowles, 1995; Schnell and King, 1994; Bosse et al., 1993; Nold et al., 1999), was not included in our model; this might need to be improved in future work. The shortage of field observation has long been identified as one of the biases in summarization for scientific induction (Tian et al., 1998; Schimel et al., 2000). The different methods used in our study and Liu and Greaver's study might explain the difference between two studies; our study actually cover all the area of same biome type across
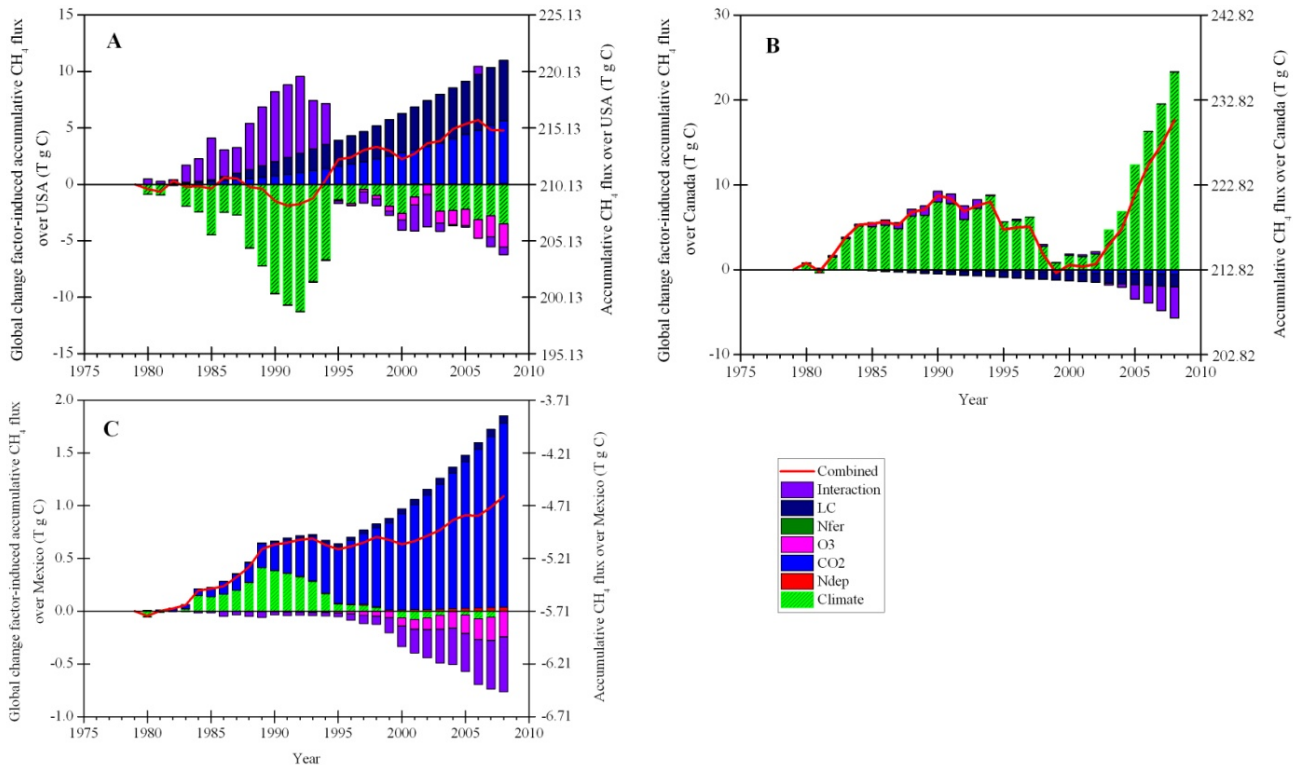
**Fig. 7.** Factorial contributions to accumulated CH$_4$ flux at country-level during 1979–2008 ((**A**): USA; (**B**): Canada; (**C**): Mexico) (The right Y-axis shows the accumulated CH$_4$ flux with baseline; Interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; Nfer means contribution from N fertilizer application; O$_3$ means contribution from O$_3$ pollution; CO$_2$ means contribution from elevated atmospheric CO$_2$; Ndep means contribution from N deposition; Climate means contribution from climate variability).
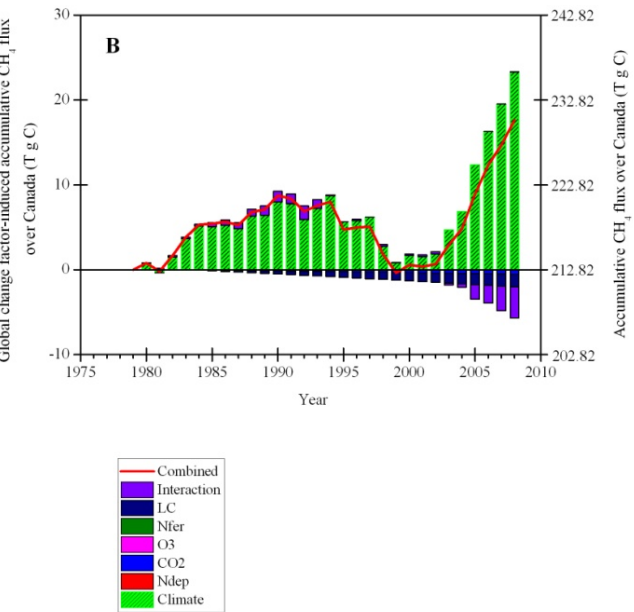


**Fig. 8.** Factorial contribution to the inter-annual variations in CH$_4$ flux over North America (The right Y-axis shows the accumulated CH$_4$ flux with baseline; Interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; Nfer means contribution from N fertilizer application; O$_3$ means contribution from O$_3$ pollution; CO$_2$ means contribution from elevated atmospheric CO$_2$; Ndep means contribution from N deposition; Climate means contribution from climate variability).

North America, while Liu and Greaver's study only contain few data points across the globe, even rarer for North America. Given the large CH$_4$ flux and N limitation for most of the wetland ecosystems (LeBauer and Treseder, 2008; Morris, 1991), a small amount of N input might significantly stimulate CH$_4$ emission (Zhang et al., 2007b). DLEM-estimated N input effect on CH$_4$ emission in wetlands is $272 \pm 15$ mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$ compared to $8 \pm 4$ mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$ in Liu and Greaver's study (2009) and 676 mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$ in a field experiment (Zhang et al., 2007c). The effects from climate variability and land conversion are more dependent on driving data; we assumed our results are reliable in simulating effects of land conversion and climate change on CH$_4$ flux as our model works fairly well in estimating absolute flux of CH$_4$ in most biomes in response to climate variability and other driving forces (Tian et al., 2010a).

Model-estimated N deposition-induced CH$_4$ uptake is $-0.21 \pm 0.02$ mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$ for grassland comparing to 0 mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$ in Liu and Greaver's study (2009). DLEM-estimated decrease in CH$_4$ uptake in response to N input is due to N induced decrease in CH$_4$ oxidation (Nold et al., 1999). The reported null
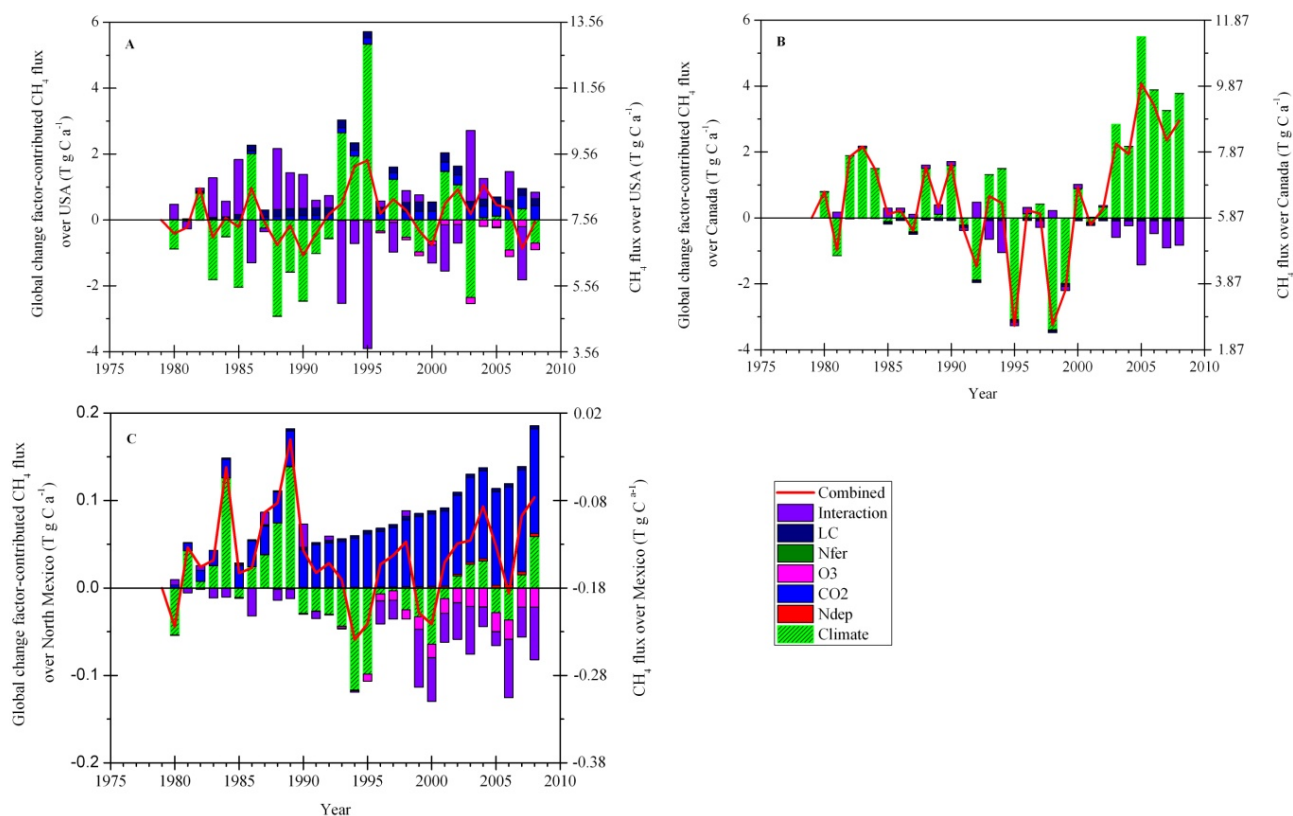
**Fig. 9.** Factorial contribution to the inter-annual variations in CH$_4$ flux by country (**(A)**: USA; **(B)**: Canada; **(C)**: Mexico) (The right Y-axis shows the accumulated CH$_4$ flux with baseline; Interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; Nfer means contribution from N fertilizer application; O$_3$ means contribution from O$_3$ pollution; CO$_2$ means contribution from elevated atmospheric CO$_2$; Ndep means contribution from N deposition; Climate means contribution from climate variability).

response of CH$_4$ flux in grassland in response to N input in Liu and Greaver's study might be due to lack of observations (2009).

## 4.2 Factorial controls on CH$_4$ flux

The enhancements of CH$_4$ emission by N input, including atmospheric deposition and anthropogenic fertilizer application, and elevated atmospheric CO$_2$ concentration are possibly due to the higher substrate caused by higher net primary production in response to elevated atmospheric CO$_2$ and N input (Magnani et al., 2007; Reich et al., 2001, 2006); the continental-average N deposition has increased from 0.28 g N m$^{-2}$ a$^{-1}$ in 1979 to 0.39 g N m$^{-2}$ a$^{-1}$ in 2008; and N fertilizer application rate has increased from 4.92 g N m$^{-2}$ a$^{-1}$ in 1979 to 6.92 g N m$^{-2}$ a$^{-1}$ in 2007; O$_3$ pollution decreased CH$_4$ emission over North America, in the USA and Canada which is probably due to the negative effect posed by O$_3$ on plant (Morsky et al., 2008). The effects of land conversion on CH$_4$ emission really depends on the direction of land conversion, if the conversion is from wetland to other ecosystem types, the CH$_4$ emission will definitely decrease (Inubushi et al., 2003; Jiang et al., 2009).

## 4.3 Inter-annual variability in CH$_4$ flux

The overall increases in terrestrial CH$_4$ emission over North America caused by global change factors could be primarily attributed to climate variability during 1979–2008 (Fig. 6). This indicates a potential increase in atmospheric CH$_4$ concentration resulted from accelerating CH$_4$ emission from terrestrial ecosystem under the future climate change projected by many general circulation models (Forster et al., 2007).

The inter-annual variability in the continental CH$_4$ flux was dominated by climatic variability (Table 3); this would be supported by the significantly positive correlation between climate-induced and overall CH$_4$ fluxes (Fig. 4), and the detailed analysis of factorial contribution to terrestrial CH$_4$ flux over the 30 years (Fig. 6). Meanwhile, the long-term trend of CH$_4$ flux was also contributed from rising atmospheric CO$_2$ concentration, N deposition, O$_3$ pollution, N fertilizer application, and land conversion. The climate variability increased CH$_4$ emission from North American terrestrial ecosystems; this is primarily resulted from the climatic effects on CH$_4$ emission over Canada. The increased temperature are primarily occurred in Canada, given that the temperature sensitivity of soil organic matter decomposition is

**Table 6.** Comparison of factorial effects on CH$_4$ fluxes against other studies (positive values mean increase, while negative values mean decrease, either in CH$_4$ uptake or in CH$_4$ emission).

| | Biome | Experiment design | This study | Others | Reference |
|---|---|---|---|---|---|
| Elevated CO$_2$ concentration | Mire, wetland | Double CO$_2$ or 200 ppm increase from 355 ppm to 550 ppm | +58% in CH$_4$ emission* | 0 ~ +146% in CH$_4$ emission | Saarnio and Silvola (1999); Megonigal and Schlesinger (1997); Cheng et al. (2006); Dacey et al. (1994); Saarnio et al. (1998); Silvola et al. (2003); Vann and Megonigal (2003); Hutchin et al. (1995) |
| | Temperate forest | 360 ppm rose to 560 ppm of atmospheric CO$_2$ | −3% in CH$_4$ consumption** | −9 ~ −30% in CH$_4$ consumption | Phillips et al. (2001); Ambus and Robertson (1999) |
| | meadow | +100 ppm increase on ambient CO$_2$ | −1% in CH$_4$ consumption*** | Negative yet not significant in CH$_4$ consumption | Kanerva et al. (2007) |
| N input | Forest | Meta-analysis | −0.32 ± 0.02 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | 17 ± 5 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | Liu and Greaver (2009) |
| | Forest | Field experiment with 0, 3.7 and 12 g N m$^{-2}$ a$^{-1}$ application | | −10.75 ± 3.98 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake**** | Steudler et al. (1989) |
| | Wetland | Meta-analysis | 272 ± 15 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ emission | 8 ± 4 (mg C g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ emission | Liu and Greaver (2009) |
| | Wetland | Field experiment with 0 and 24 g N m$^{-2}$ a$^{-1}$ application | | 676 (mg C g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ emission***** | Zhang et al. (2007c) |
| | Grassland | Meta-analysis | −0.21 ± 0.02 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | 0 (mg C g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | Liu and Greaver (2009) |
| | Dry cropland | Meta-analysis | 7.43 ± 1.09 (mg C m$^{-2}$ a$^{-1}$ per g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | 12 ± 6 (mg C g N$^{-1}$ m$^{-2}$ a$^{-1}$) in CH$_4$ uptake | Liu and Greaver (2009) |
| O$_3$ pollution | Peat land | Double ambient O$_3$ | Negative yet not significant in CH$_4$ emission | Negative yet not significant in CH$_4$ emission | Morsky et al. (2008) |
| | meadow | 10–20 ppb higher than ambient | Negative yet not significant in CH$_4$ uptake | Negative yet not significant in CH$_4$ uptake | Kanerva et al. (2007) |

* The value is estimated by the linear calculation based on regressed equation between atmospheric CO$_2$ concentration (ppm) and annual CH$_4$ emission from herbaceous wetland over North America ($Y = 6.82 * X + 4754.6$, $R^2 = 0.996$, $N = 30$).
** The value is estimated by the linearly calculation based on regressed equation between atmospheric CO$_2$ concentration (ppm) and annual CH$_4$ emission from forests over North America ($Y = 0.01 * X - 158.92$, $R^2 = 0.99$, $N = 30$).
*** The value is estimated by the linearly calculation based on regressed equation between atmospheric CO$_2$ concentration (ppm) and annual CH$_4$ emission from grassland over North America ($Y = 0.05 * X - 568.82$, $R^2 = 0.96$, $N = 30$).
**** Averaged for hardwood and pine temperate forest from the field experimental results with 200 days of frost-free days.
***** Calculated from the field experimental results in May, June, July, August the growing season of wetland vegetation.

The effects of N input were summarized based on meta-analysis in Liu and Greaver's study (2009); the effects in this study were calculated based on N deposition-induced changes in CH$_4$ flux for forest, grassland, and wetland, and N fertilizer-induced changes in CH$_4$ flux for dry cropland.

higher in high-latitudinal Canada than those in mid and low latitudinal US and Mexico (Davidson and Janssens, 2006), the increased temperature possibly leads to more DOC in Canada which is the substrate of CH$_4$ production and finally leads to higher CH$_4$ emission. This is consistent with previous studies (Zhuang et al., 2004, 2006). The increase in terrestrial CH$_4$ flux over North America during 2005–2007 is primarily attributable to climate variability (Fig. 8); the increases in CH$_4$ emission is consistent with the increase in atmospheric CH$_4$ concentration in 2007 (Rigby et al., 2008; Dlugokencky et al., 2009), suggesting that the newly-found increase in atmospheric CH$_4$ concentration in 2007 might be caused by global environment change, especially climate variability.

The contrasting effects of climate variability from 1979 to 2008 on the CH$_4$ emissions from USA and Canada may be due to the different ecosystem responses to interactions among climate variables (Table 5). As reported that higher increases in air temperature and precipitation occur in

Canada than in USA (Groisman and Easterling, 1994; Christensen et al., 2007), which may lead to more substrate and more CH$_4$ production and higher CH$_4$ emission; this is consistent with a number of field observations (Schrope et al., 1999; Song et al., 2009). However, it should be noted that the single climate variables might played contrasting role in affecting terrestrial CH$_4$ flux. For example, the temperature effect on CH$_4$ emission is positive in the USA, yet negative in the Canada; while the effect of multiple factors interaction is positive in Canada, yet negative in the USA (Table 5).

## 4.4 Interactions among multiple factors

Through this study, we also found that the interactive effects among global change factors played an important role in contributing to terrestrial CH$_4$ flux. The interaction among global change factors has been recognized long before (Dermody, 2006); most of the field experiment still treat it as negligible, although few experiments have introduced this in

their experiment design (Xia et al., 2009; Reich et al., 2006). The interactive effects among more than three factors are still short of investigation (Heimann and Reichstein, 2008). This study shows that the modeling approach may serve as one complementary tool for the field experiments in addressing interactive effect among multiple factors.

## 4.5 Uncertainties

This study examined the factorial contributions to temporal and spatial variations in CH$_4$ flux over North American terrestrial ecosystems during 1979–2008. There are several uncertainties which need to be eliminated in our future work. First, the climate data used in this study only cover the time period of 1979–2008; the legacy effects of the pre-1979 global change factors could not be included in this study; this might overestimate or underestimate the long-term accumulated CH$_4$ flux. Second, most of the single factor effects on CH$_4$ flux have not been fully validated because of the scarcity of the field experiments (Heimann and Reichstein, 2008). Third, some possible disturbances or environmental factors probably influencing CH$_4$ flux were not included in this study; for example, the fire (Burke et al., 1997), thaw-freezing cycle in high-latitudinal ecosystems (Turetsky and Louis, 2006; Mastepanov et al., 2008), and insect outbreak (Turetsky and Louis, 2006); all these factors will be important but challenging to be included in the process-based modeling approach. Fourth, the open water emission of CH$_4$ is a globally significant CH$_4$ source (Bastviken et al., 2004; Walter et al., 2006, 2007), which may contribute to the terrestrial CH$_4$ budget, especially from inland small lakes or river (Walter et al., 2006, 2007). Fifth, the uncertainties caused by model structure, parameters, and input data might need to be evaluated for accurately quantifying the relative contribution of each factor to the regional CH$_4$ flux. Last but not least, the mechanisms for CH$_4$ flux in response to global change factors need to be improved in future studies, as the global change factors may yield different impacts on production and consumption of atmospheric CH$_4$. Partitioning the effects of global change factors on CH$_4$ production and consumption may be one of the major efforts improving our estimation of regional CH$_4$ flux in the context of changing environment.

## 5 Conclusions

Factorial contributions to the spatial and temporal variations in CH$_4$ flux over North America were examined at both continental and country levels by using a highly integrated process-based model driven by multiple global change factors including changing climate, N deposition, rising atmospheric CO$_2$, O$_3$ pollution, N fertilizer application, and land conversion. Although some uncertainties, the attribution of spatial and temporal variations in CH$_4$ flux over North America to six factors and their interaction is helpful in advancing

our understanding of the dynamics of atmospheric CH$_4$ concentration; it might also benefit the policy-making for curbing the increase in atmospheric CH$_4$ concentration. This study found the contrasting climatic effects on CH$_4$ emissions from the USA and Canada. The complicated effects of multiple-factor interaction on CH$_4$ flux suggest that the current experiments which usually ignore the interactive effects from multiple-factor may lead to biases in the estimation of CH$_4$ flux. This study also pointed out that the models driven by few global change factors may bring bias in estimating CH$_4$ flux. The climate-dominated inter-annual variations in CH$_4$ flux pretends a changed regime of CH$_4$ exchange between terrestrial ecosystems and the atmosphere in the response to projected climate change (Forster et al., 2007).

This study also provides insights for the examination of multiple-factor interactive effects on terrestrial CH$_4$ flux. Given the advantages of modeling approach in quantifying regional CH$_4$ flux and the importance of field experiments in model improvement and flux estimation, clearly, a collaborative effort between field ecologists and modelers is necessary for further investigation of the underlying mechanisms for spatial and temporal variations in CH$_4$ exchange between terrestrial ecosystems and the atmosphere.

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