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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 factors and their interaction. Over the past three decades, our simulation indicates that global change factors accumulatively contributed 43.05 Tg 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 2.69 Tg CH_4-C, all other factors including climate variability, nitrogen (N) deposition, rising atmospheric carbon dioxide (CO_2), N fertilization, and land conversion increased terrestrial CH_4 emissions by 40.37 Tg CH_4-C, 0.42 Tg CH_4-C, 6.95 Tg CH_4-C, 0.11 Tg CH_4-C, and 3.70 Tg CH_4-C, respectively, and interaction between/among these global change factors led to a decline of CH_4 emission by 5.80 Tg CH_4-C. Climatic variability dominated the inter-annual variations in terrestrial CH_4 fluxes at both continental and country levels. The relative importance of each environmental factor in determining the magnitude of methane flux shows substantially spatial variation across North America. This factorial attribution of CH_4 fluxes over the 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,

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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 estimation (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 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 "real 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, consumption, and transport from soil/water to the atmosphere 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 flux by stimulating CH_4 emission (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

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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 simultaneously 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

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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 type, and N fertilization. The interactive effects among these six factors were calculated by subtracting the changes in CH_4 flux contributed from all factors by the changes in CH_4 flux caused by six individual factors (see Sect. 2.3).

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., 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, fertilization, rotation, and harvest can

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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., 2010a), 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 include ebullition, diffusion, and plant-mediated transport, are considered in the DLEM (Tian et al., 2008, 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.

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

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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 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; $f(T, WFPS)$ 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

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deposition rate, annual land-cover change and land management practices (including fertilization, irrigation) for the entire North America. The climate dataset was generated based on NARR dataset. 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). O_3 pollution data was retrieved from a global dataset developed by Felzer et al. (Felzer et al., 2005), the time period covers 1900–2050. 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). Fertilization 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 reprojected to a consistent 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 from 1979–2008; 2) historical annual N deposition from 1901 to 2008; 3)

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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 unchanged due to shortage of data; and 6) historical N fertilization 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 fertilization on the terrestrial CH_4 flux over North America, we conducted seven simulations in this study. 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 fertilization 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 fertilization data were kept constant in the year of 1900.

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

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$<0.1\,g\,m^{-2}$ for N storage). After the system reaches 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 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 model (McGuire et al., 2001). Baseline flux was defined as the CH_4 flux in 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 in this study.

2.5 Statistical analysis

The regression analysis was used in this study to find the long-term changing trend 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.

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 1, 2). For the climatic variables, maximum, minimum, and average temperatures, and solar radiation showed significantly

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increasing trends at the rates of 0.04±0.01 °C a^{-1}, 0.03±0.01 °C a^{-1}, 0.03±0.01 °C a^{-1}, and 0.17±0.03 W m^{-2} a^{-1}, respectively; yet precipitation and relative humidity did not show any significantly changing trends along the study period. All the other driving factors significantly increased since 1979; the long-term increasing rates were 40±10 ppb-h a^{-1} for O_3 pollution, 1.98±0.12 mg m^{-2} a^{-1} for N deposition, 0.06±0.01 g m^{-2} a^{-1} for N fertilization, and 1.66±0.02 ppm a^{-1} for atmospheric CO_2 concentration, respectively. The area of different land cover types changed slightly through 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 completely different.

Spatial variations of input data including potential vegetation distribution, N deposition, N fertilization rate, and O_3 pollution were shown in Fig. 3. The Fig. 3a shows the contemporary spatial distribution of vegetation used in this study; it should be noted that natural wetlands primarily distribute in Alaska, Western Canada, south to the Hudson Bay, eastern coastal area, and Florida in the USA (Fig. 3a). The severely O_3-polluted area over North America locates in western part of North America such as the Southeastern USA which could be as high as more than 5000 ppb h^{-1} (monthly accumulated hourly O_3 dose over a threshold of 40 ppb in ppb-h), while the other areas, especially northern end of continental North America, feature low O_3 pollution (Fig. 3b). The major cropland with high N fertilizer application (larger than 10 g N m^{-2} a^{-1}) locate in USA, including western, central, and eastern costal area of USA. The Canada and Mexico had small amount of cropland and received lower application rate of N fertilizer (Fig. 3c). 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. 3d).

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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. 4); 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 continental North America, including southern USA and most of Mexico; and other areas acted as a weak sink of atmospheric CH_4.

Various factors did not exert substantially different effects on the spatial distribution of CH_4 flux across the entire continent except central part of North America (Fig. 4). Without land use change, the central part of North America acted as a strong source of CH_4 (Fig. 4a–e); while it acted as a sink of CH_4 when land use change was introduced (Fig. 4f and g).

3.3 Temporal patterns of CH_4 flux from 1979 to 2008

The CH_4 flux over the entire continental North America showed significant inter-annual fluctuations from 1979 to 2008 (Fig. 5). The lowest annual CH_4 emission was 11.74 Tg CH_4-C a^{-1} in 1998, and the highest was 18.42 Tg CH_4-C a^{-1} in 2005. Before 2001, the CH_4 flux did not show any significantly changing trend; 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±1.64 Tg CH_4-C a^{-1}; and the overall increasing rate of CH_4 flux was 0.10 Tg CH_4-C a^{-1} over study period (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

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0.15 ± 0.04 Tg 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 Tg CH_4-C a^{-1} (P<0.001) (Fig. 5d), while O_3 pollution continuously decreased CH_4 emission at a rate of 0.01 ± 0.001 Tg CH_4-C a^{-1} (P<0.001) (Fig. 5e). N deposition generated an increasing rate of 0.001 Tg CH_4-C a^{-1} to continental-level CH_4 emission (P<0.001) (Fig. 5c), while N fertilization 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, an significantly increasing trend at a rate of 0.007 ± 0.001 Tg 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).

3.4 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 in 1979 is the baseline emission, and the changes in CH_4 flux compared to the year of 1979 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 of 1979, 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, eastern parts of Canada, Northwestern and Northeastern USA, and portions of Mexico, while decreased CH_4 emission in central USA and portions of Alaska (Fig. 6a); N deposition enhanced CH_4 emission in Eastern

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USA and portions of Northwestern Canada (Fig. 6b); elevated atmospheric CO_2 enhanced CH_4 emission over large area of continental North America, but did not affect CH_4 flux in Western USA and majority of Mexico (Fig. 6c); O_3 pollution exerted no significant effects on CH_4 flux across majority of North America, while decreased CH_4 emission in eastern continental North America and enhanced CH_4 emission in small magnitude over portions of Canada (Fig. 6d); N fertilization and land conversion slightly enhanced CH_4 emission in agricultural land throughout North America (Fig. 6e and f); interactive effects between/among global change factors enhanced CH_4 emission in large area of North America, especially Northwest (Fig. 6h); combining all the effects from various global change factors, the CH_4 emission was enhanced across majority of the continental North America over the past three decades (Fig. 6g).

3.5 Factorial contributions to the accumulated CH_4 flux over North America during 1979–2008

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. Through the 30-year study period, the accumulated continental CH_4 flux over North America was 440.75 Tg CH_4-C, of which 397.70 Tg CH_4-C was contributed from baseline flux and 43.05 Tg CH_4-C was caused by global change factors (Table 3). O_3 pollution and the interactive effects between/among multiple factors decreased CH_4 emission by 2.70 Tg CH_4-C and 5.80 Tg CH_4-C, respectively, while all the other single factors increased CH_4 emission from North America's terrestrial ecosystems. Climate variability, N deposition, elevated atmospheric CO_2, N fertilization, and land conversion enhanced continental CH_4 emission by 40.05 Tg CH_4-C, 0.42 Tg CH_4-C, 6.95 Tg CH_4-C, 0.11 Tg CH_4-C, and 3.70 Tg CH_4-C, respectively.

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For the accumulated CH_4 fluxes during 1979–2008, the baseline emission, climate variability, N deposition, elevated atmospheric CO_2, O_3 pollution, N fertilization, land conversion, and multiple-factor interaction contributed 90.23%, 9.16%, 0.16%, 1.58%, −0.61%, 0.02%, 0.84%, and −1.32%, respectively, to the continental-level emission over the entire continental North America (Table 3).

3.6 Factorial contributions to the accumulated CH_4 flux at country-level during 1979–2008

The 30-year accumulated CH_4 emission was 214.89 Tg CH_4-C for USA and 230.47 Tg CH_4-C for Canada, respectively. Mexico acted as a sink for atmospheric CH_4, and the total sink strength was 5.28 Tg CH_4-C over the past 30 years (Table 3). For USA, climate variability and O_3 pollution substantially decreased CH_4 emission by 20.45 Tg CH_4-C and 2.51 Tg CH_4-C, respectively, during 1979–2008, while N deposition, elevated atmospheric CO_2 and N fertilization, and land conversion enhanced CH_4 emissions by 0.28 Tg CH_4-C, 5.91 Tg CH_4-C, 0.06 Tg CH_4-C, and 4.61 Tg CH_4-C, respectively (Table 3). After removing the baseline emission in 1979, the global change factors decreased CH_4 emission from USA's terrestrial ecosystems during 1979–2008. For Canada, climate variability significantly enhanced CH_4 emission by 61.49 Tg CH_4-C during 1979–2008, N deposition, O_3 pollution, and N fertilization increased CH_4 emissions by 0.05 Tg CH_4-C, 0.03 Tg CH_4-C, 0.03 Tg CH_4-C, respectively, for the same period; while elevated atmospheric CO_2, land conversion and multiple-factor interaction decreased CH_4 emission by 0.76 Tg CH_4-C, 1.01 Tg CH_4-C, and 5.44 Tg CH_4-C, respectively (Table 3). After removing the baseline emission in 1979, the global change factors increased CH_4 emission from Canada's terrestrial ecosystems during 1979–2008. For Mexico, climate variability, O_3 pollution, and multiple-factor interaction enhanced CH_4 consumption by 0.67 Tg CH_4-C, 0.21 Tg CH_4-C, and 0.46 Tg CH_4-C, respectively, for 1979–2008; while N deposition, elevated atmospheric CO_2, N fertilization, and land conversion decreased CH_4 consumption by 0.10 T g CH_4-C, 1.80 Tg CH_4-C, 0.01 Tg CH_4-C, and 0.1 Tg CH_4-C, respectively, for the same

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time period (Table 3). After removing the baseline flux in 1979, the global change factors increased CH_4 uptake from Mexico's terrestrial ecosystems during 1979–2008.

For the country-level accumulated CH_4 fluxes over 30 years, the baseline emission, climate variability, N deposition, elevated atmospheric CO_2, O_3 pollution, N fertilization, land conversion, and multiple-factor interaction contributed 105.59%, −9.52%, 0.13%, 2.75%, −1.17%, 0.03%, 2.14%, and 0.04%, respectively, to the country-level emission in USA, 76.40%, 26.68%, 0.02%, −0.33%, 0.01%, 0.01%, −0.44%, and −2.36%, respectively, to the country-level emission in Canada, and 114.47%, 14.54%, −2.11%, −38.90%, 4.63%, −0.32%, −2.19%, and 9.88%, respectively, to the country-level emission in Mexico (Table 3).

3.7 Factorial contributions to the inter-annual variations in CH_4 flux over North America during 1979–2008

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 from 1979 to 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. 9). 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. 9). Climate variability-induced effects dominated the increases in CH_4 emission over three time periods: 1979–1984, 1993–1998, and 2002–2008. 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.

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3.8 Factorial contributions to the inter-annual variations in CH_4 flux at country level during 1979–2008

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 1979–1985, 1987–1990, 2000–2004, and 2006–2008; multiple-factor interaction dominated the sink of atmospheric CH_4 over USA during the time period of 1993–1999 (Fig. 10a). Climate variability outweighed other factors in controlling the increases in terrestrial CH_4 emission over Canada (Fig. 10b). Climate variability and interactive effect of multiple-factor affected the inter-annual fluctuations in terrestrial CH_4 flux over Mexico; since 1994, 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. 10c).

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 and 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

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in northwestern continental North America is due to the dense distribution of natural wetland in that region (Fig. 3a) (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 atmospheric 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 4). 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 previously reported range of 0∼146%, and is a 1% decrease in CH_4 consumption for meadow grassland, which is comparable to Kanerva et al.'s (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∼30% 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 filed 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 with previous studies; both agreed that the O_3 pollution exerted negative yet not significant effects on CH_4 from peat-land and meadow grassland (Table 4).

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 $7.43\pm1.09\,mg\,C\,m^{-2}\,a^{-1}$ per $g\,N^{-1}\,m^{-2}\,a^{-1}$ for dry cropland, comparing to $12\pm6\,mg\,C\,m^{-2}\,a^{-1}$ per $g\,N^{-1}\,m^{-2}\,a^{-1}$ in Liu and Greaver's (2009) study. 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 $-0.32\pm0.02\,mg\,C\,m^{-2}\,a^{-1}$ per $g\,N^{-1}\,m^{-2}\,a^{-1}$ for

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forest compared to 17±5 mg C m^{-2} a^{-1} per g N^{-1} m^{-2} a^{-1} in Liu and Greaver's study (2009), and −10.75±3.98 (mg C m^{-2} a^{-1} per 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 are resulted from net changes in both CH_4 production and consumption; for example the increases in CH_4 emission might be resulted from either increases in CH_4 production or decreases in CH_4 consumption; the increases in CH_4 uptake might be resulted from either increases in CH_4 oxidation or decreases in CH_4 production; Liu and Greaver's (2009) study solely reported production or uptake, while this study reported the net flux from production, oxidation, and transport (Sect. 2.1).

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 North America, while Liu and Greaver's study only contain few data points across globe, even rarer for North America. Given the large CH_4 flux and N limitation for most of 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 wetland is 272±15 mg C m^{-2} a^{-1} per g N^{-1} m^{-2} a^{-1} compared to 8±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

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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 ± 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 (2009) study. 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 response of CH_4 flux in grassland in response to N input in Liu and Greaver's (2009) study might be due to lack of observations.

4.2 Factorial controls on CH_4 flux

The enhancements of CH_4 emission by N input, including atmospheric deposition and anthropogenic fertilization, 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. 7). This indicates a potential increase in atmospheric CH_4 concentration resulted

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from accelerating CH_4 emission from terrestrial ecosystem under the future climate change projected by many global 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. 5), and the detailed analysis of factorial contribution to terrestrial CH_4 flux over the 30 years (Fig. 7). Meanwhile, the long-term trend of CH_4 fluxes was also contributed from rising atmospheric CO_2 concentration, N deposition, O_3 pollution, N fertilization, and land conversion. The climate variability increased CH_4 emission from North America's 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 higher in high-latitudinal Canada than those in mid and low latitudinal US and Mexico (Davidson and Janssens, 2006), the increased temperature possibly lead to more DOC in Canada which is the substrate of CH_4 production and finally lead 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. 9); the increases in CH_4 emission is consistent with the increases in atmospheric CH_4 concentration in 2007 (Rigby et al., 2008; Dlugokencky et al., 2009), suggesting that the newly-found increases in atmospheric CH_4 concentration in 2007 might be caused by global change environment, 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 elevated temperature. 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).

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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 America's terrestrial ecosystems during 1979–2008. There are several uncertainties which need to be improved 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 may influence 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., 2006a, 2007a), which may contribute to the terrestrial CH_4 budget, especially from inland small lake or river (Walter et al., 2006b, 2007b). Fifth, future work is needed to take into account

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the relative role of climate variables (temperature, precipitation, solar radiation) in controlling CH_4 flux. Sixth, the uncertainties caused by model structure, parameters, and input data might need to be evaluated for accurately quantify 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 study, 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 fertilization, 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).

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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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Table 1. Changing rates of driving factors for DLEM simulations (temperature including maximum, maximum, and average temperature, precipitation, short wave radiation, relative humidity, atmospheric CO_2 increase, O_3 pollution, N deposition, land use change, N fertilization).

Variables		Changing trends (Mean±SD)
Climate	Maximum temperature ($^{\circ}C\,a^{-1}$)	0.04±0.01
	Minimum temperature ($^{\circ}C\,a^{-1}$)	0.03±0.01
	Average temperature ($^{\circ}C\,a^{-1}$)	0.03±0.01
	Precipitation ($mm\,a^{-1}$)	0.65±0.65[a]
	Relative humidity ($\%\,a^{-1}$)	−0.01±0.01[a]
	Solar radiation ($W\,m^{-2}\,a^{-1}$)	0.17±0.03
Others	O_3 pollution ($ppm\text{-}h\,a^{-1}$)	0.93±0.09
	N deposition ($mg\,m^{-2}a^{-1}$)	1.98±0.12
	N fertilization ($mg\,m^{-2}a^{-1}$)	0.06±0.01
	Atmospheric CO_2 concentration ($ppm\,a^{-1}$)	1.66±0.02

a Indicates the changing trend is not significantly different from zero; positive values represent increase through the study period, and negative values represent decrease through the study period.

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Table 2. 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	6.93~6.99	3.57~3.59	2.61~2.64	2.06~2.07	0.53~0.60	2.51~2.59
Percentage	18.09	31.10	15.98	11.72	9.23	2.49	11.39

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

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Table 3. Factorial contributions to the accumulated CH_4 from 1979 to 2008 (combined represents the effects with all six factors being considered; Climate represents the impacts of climate variability only; N_{dep} represents the impacts of N deposition; CO_2 represents the impacts of CO_2 variation; O_3 represents the impacts of O_3 pollution; N_{fer} represents the impacts of N fertilization; 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).

		Baseline	Climate	N_{dep}	CO_2	O_3	N_{fer}	Land conversion	Interaction	Total flux
US	Accumulated CH_4 flux (T g C)	226.90	–20.45	0.28	5.91	–2.51	0.06	4.61	0.09	214.89
	Percentage (%)	105.59	–9.52	0.13	2.75	–1.17	0.03	2.14	0.04	100
Canada	Accumulated CH_4 flux (T g C)	176.08	61.49	0.05	–0.76	0.03	0.03	–1.01	–5.44	230.47
	Percentage (%)	76.40	26.68	0.02	–0.33	0.01	0.01	–0.44	–2.36	100
Mexico	Accumulated CH_4 flux (T g C)	–5.28	–0.67	0.10	1.80	–0.21	0.01	0.10	–0.46	–4.62
	Percentage (%)	114.47	14.54	–2.11	–38.90	4.63	–0.32	–2.19	9.88	100
NA	Accumulated CH_4 flux (T g C)	397.70	40.37	0.42	6.95	–2.69	0.11	3.70	–5.80	440.75
	Percentage (%)	90.23	9.16	0.10	1.58	–0.61	0.02	0.84	–1.32	100

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

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Table 4. 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)
		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)
		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 ($Y=6.82*X+4754.6$, $R^2=0.9963$, $N=30$).
** 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 ($Y=0.0103*X-158.92$, $R^2=0.9902$, $N=30$).
*** 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 ($Y=0.047*X-568.82$, $R^2=0.959$, $N=30$).
€ Correspondingly in line nitrogen input on forest (Steudler et al., 1989). Averaged for hardwood and pine temperate forest from the field experimental results with 200 days of frost-free days.
£ correspondingly in line nitrogen input effects on wetland (Zhang et al., 2007). 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.

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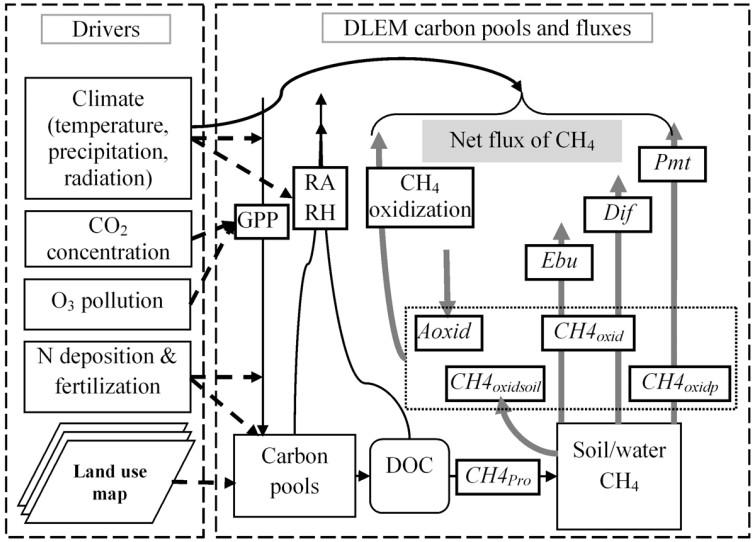

Fig. 1. Conceptual diagram showing major processes for CH_4 production, oxidation and transport from the soil/water to the atmosphere in response to multiple global change factors.

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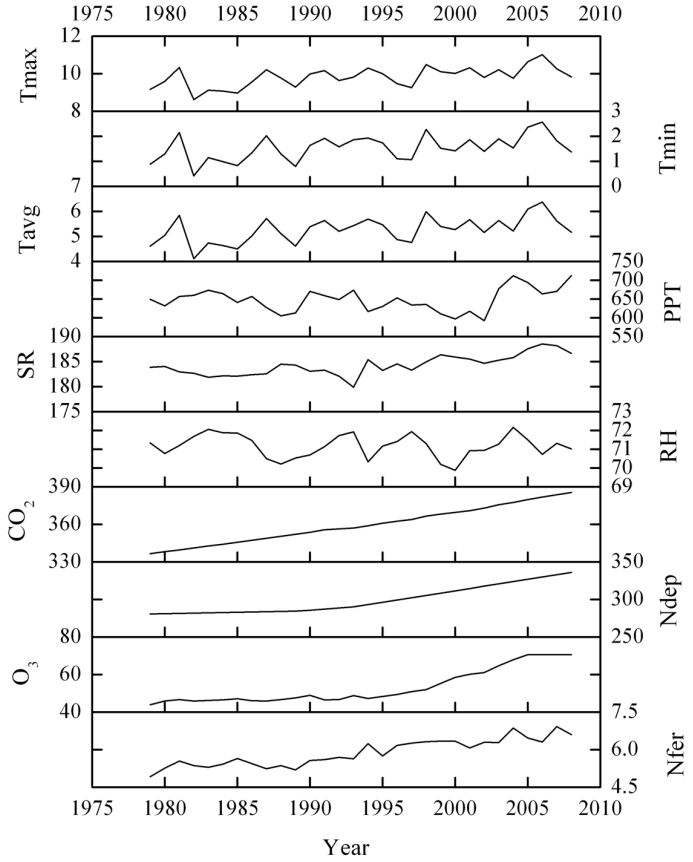

Fig. 2. Temporal dynamics of the variables from 1979 to 2008 (T_{max}: maximum temperature (°C); T_{min}: minimum temperature (°C); T_{avg}: average temperature (°C); PPT: precipitation (mm); SR: solar radiation ($W\,m^{-2}$); RH: relative humidity (%); CO_2: atmospheric CO_2 concentration (ppm); N_{dep}: N deposition ($mg\,N\,m^{-2}$); O_3: O_3 pollution (ppb-h); N_{fer}: N fertilization ($g\,N\,m^{-2}$)).

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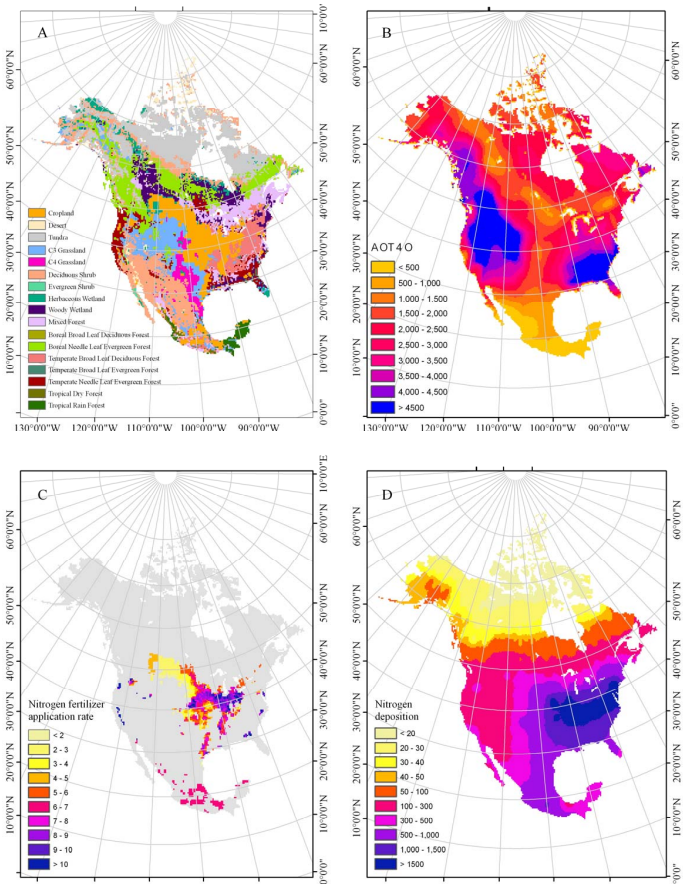

Fig. 3. (A) Contemporary vegetation map, and spatial distribution of 30-year averages of **(B)** O_3 pollution, **(C)** N fertilization application, and **(D)** N deposition rate (unit of O_3 is: AOT40 at ppb/h; Unit of N fertilization application rate is g N m^{-2} a^{-1}; Unit of N deposition is: mg N m^{-2} a^{-1}).

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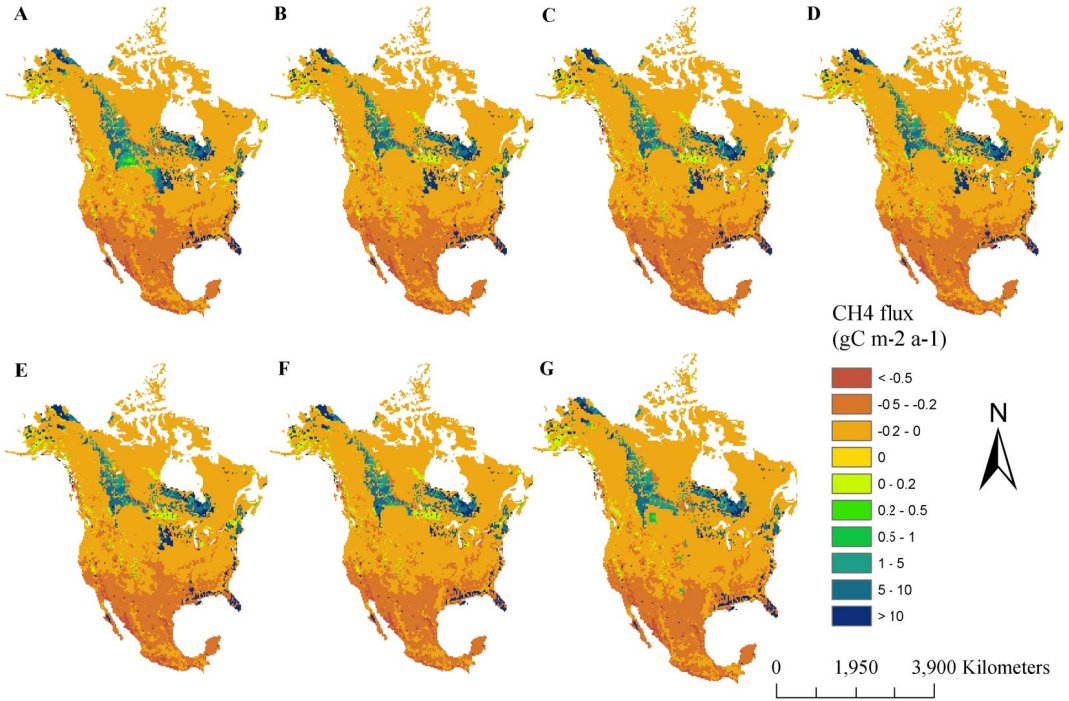

Fig. 4. Spatial variations of terrestrial CH_4 fluxes caused by global change factors over North America from 1979 to 2008 – **(A)**: climatic variability; **(B)**: N deposition; **(C)**: CO_2; **(D)**: O_3 pollution; **(E)**: N fertilization; **(F)**: land conversion; **(G)**: all combined.

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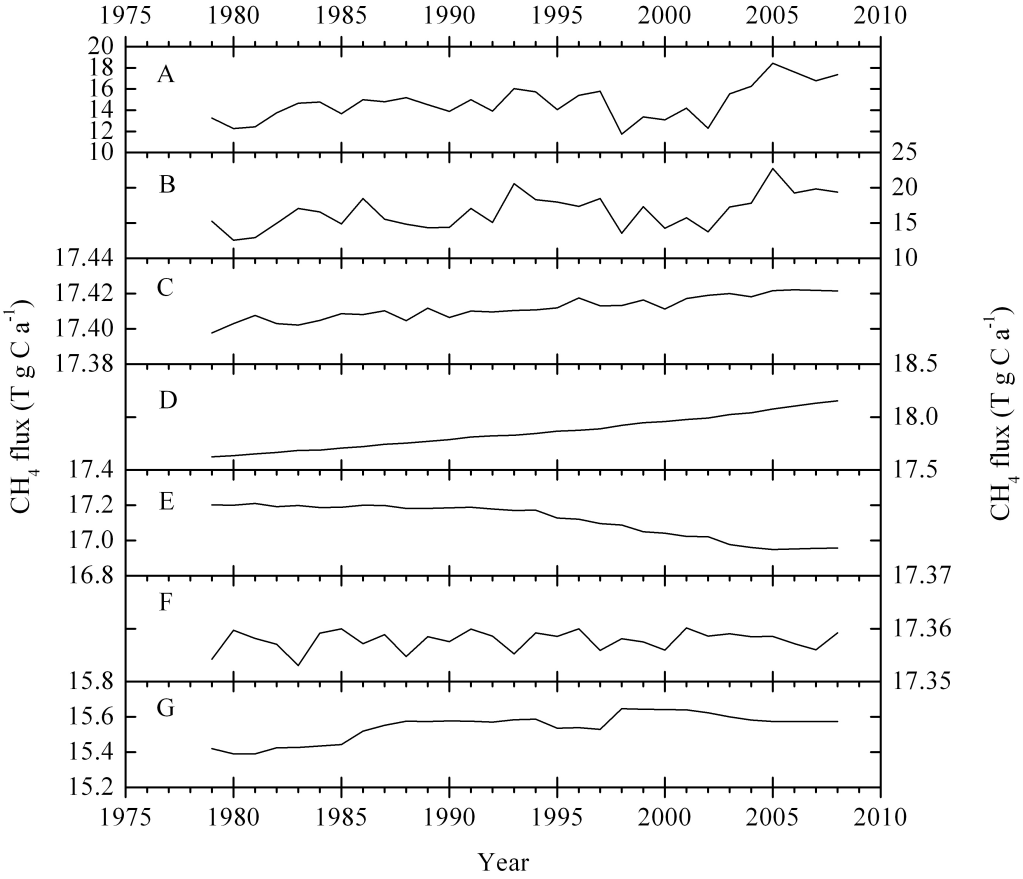

Fig. 5. Temporal variations of terrestrial CH_4 flux caused by global change factors over North America from 1979 to 2008 – **(A)**: all combined simulation; **(B)**: climate only simulation; **(C)**: N deposition only simulation; **(D)**: CO_2 only simulation; **(E)**: O_3 only simulation; **(F)**: N fertilization simulation; **(G)**: land conversion only simulation.

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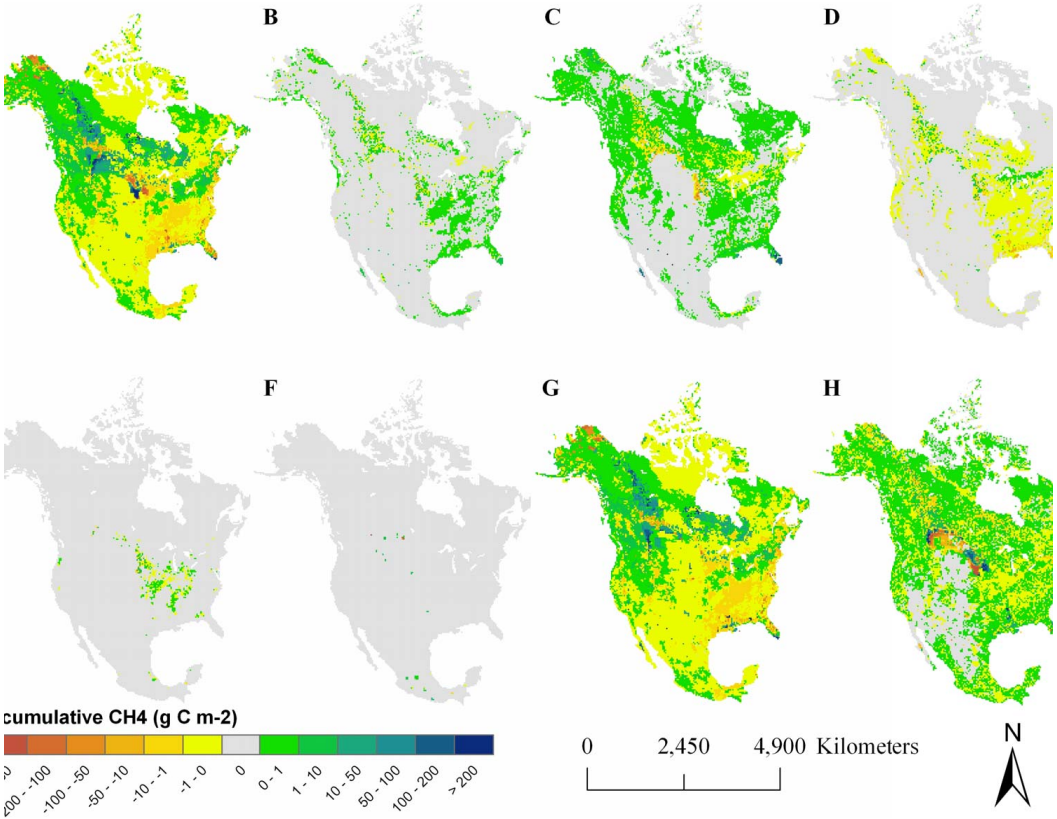

Fig. 6. Factorial contributions to the spatial variations in accumulated CH_4 flux over North America from 1979 to 2008 – **(A)**: climatic variability; **(B)**: N deposition; **(C)**: CO_2; **(D)**: O_3 pollution; **(E)**: N fertilization; **(F)**: land conversion; **(G)**: all combined; **(H)**: interaction.

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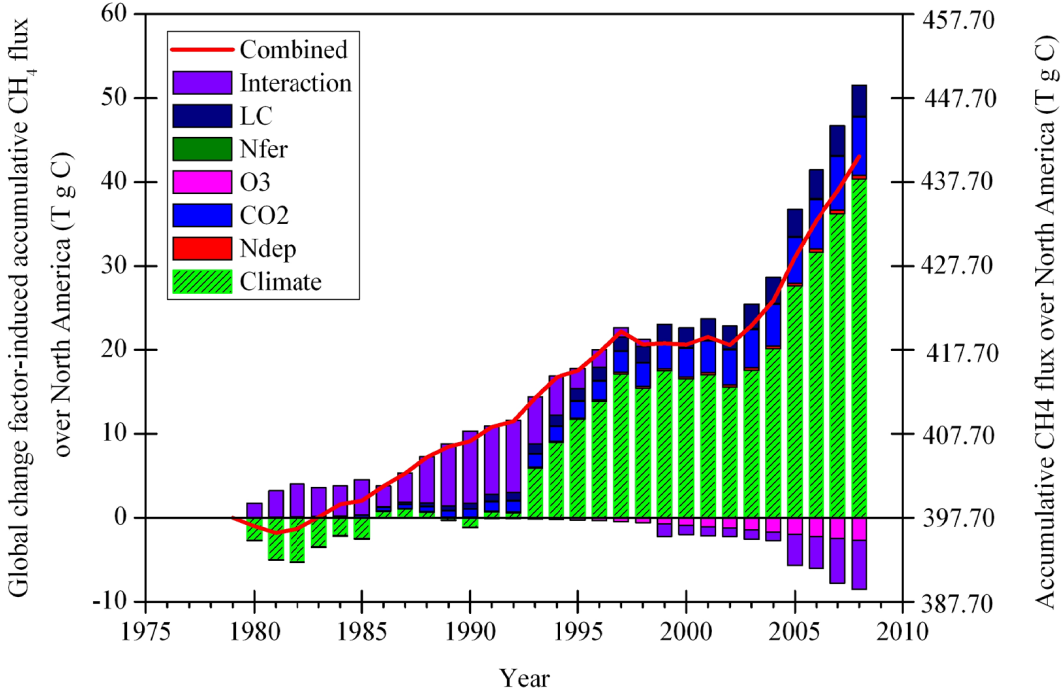

Fig. 7. Factorial contributions to accumulated CH_4 flux over North America during 1979–2008 (the right *Y*-axis shows the accumulated CH_4 flux with baseline; interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; N_{fer} means contribution from N fertilization; O_3 means contribution from O_3 pollution; CO_2 means contribution from elevated atmospheric CO_2; N_{dep} means contribution from N deposition; climate means contribution from climate variability).

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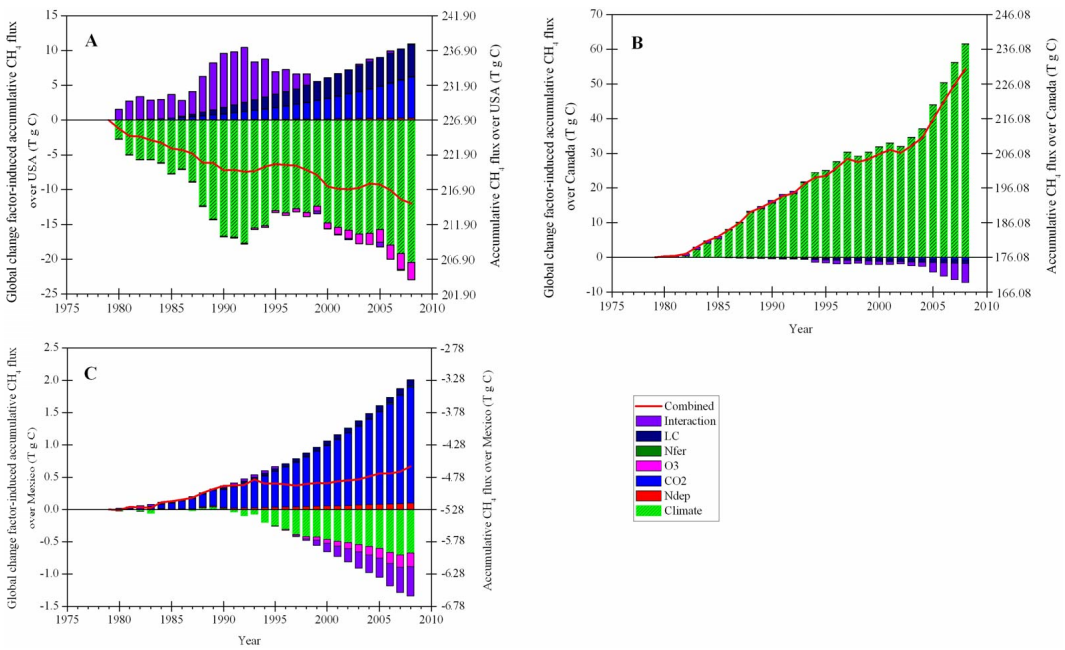

Fig. 8. Factorial contributions to accumulated CH_4 flux at country-level during 1979–2008 – **(A)**: USA; **(B)**: Canada; **(C)**: Mexico) (the right *Y*-axis shows the accumulated CH_4 flux with baseline; interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; N_{fer} means contribution from N fertilization; O_3 means contribution from O_3 pollution; CO_2 means contribution from elevated atmospheric CO_2; N_{dep} means contribution from N deposition; climate means contribution from climate variability.

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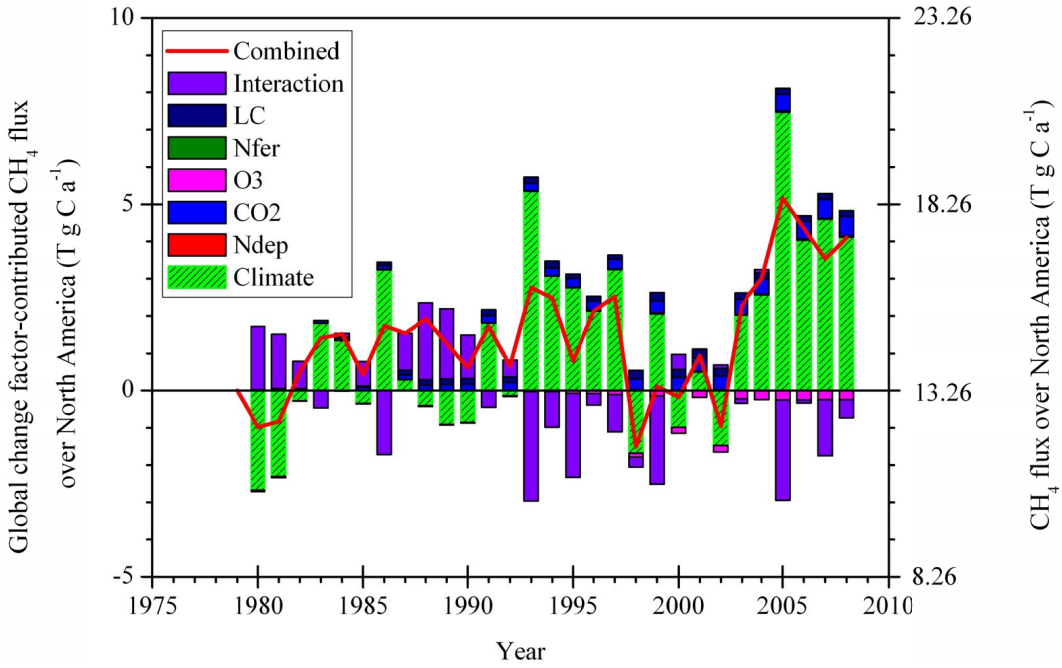

Fig. 9. Factorial contribution to the inter-annual variations in CH_4 flux over North America (the right *Y*-axis shows the accumulated CH_4 flux with baseline; interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; N_{fer} means contribution from N fertilization; O_3 means contribution from O_3 pollution; CO_2 means contribution from elevated atmospheric CO_2; N_{dep} means contribution from N deposition; climate means contribution from climate variability).

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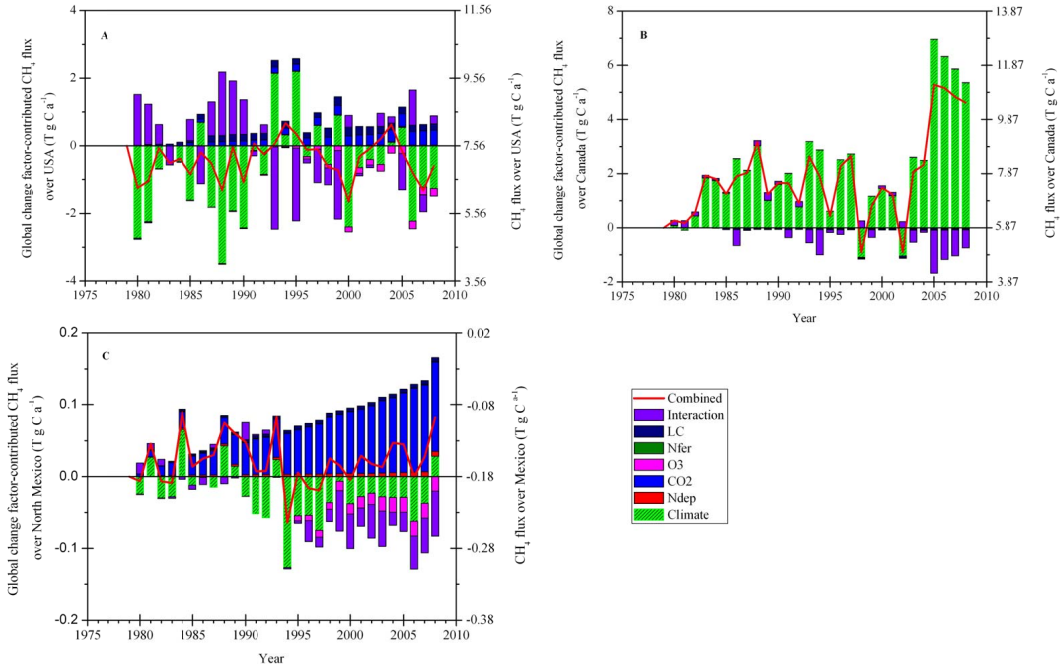

Fig. 10. Factorial contribution to the inter-annual variations in CH_4 flux by country – **(A)**: USA; **(B)**: Canada; **(C)**: Mexico) (the right *Y*-axis shows the accumulated CH_4 flux with baseline; interaction means contribution from multiple-factor interaction; LC means contribution from land conversion; N_{fer} means contribution from N fertilization; O_3 means contribution from O_3 pollution; CO_2 means contribution from elevated atmospheric CO_2; N_{dep} means contribution from N deposition; climate means contribution from climate variability.

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