Modeling Methane and Nitrous Oxide Exchanges between the Atmosphere and Terrestrial Ecosystems over North America in the Context of Multifactor Global Change

by

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Abstract

Methane (CH_4) and nitrous oxide (N_2O) are two potent greenhouse gases which in sum contribute to more than one fourth of global warming caused by anthropogenic activities. In the meantime, CH_4 and N_2O play significant roles in ozone layer chemistry. Understanding and quantifying CH_4 and N_2O fluxes in terrestrial ecosystems at large spatial scales, therefore, become an urgent task for accurately predicting climate change. In this study, I enhanced and applied a process-based ecosystem model, in conjunction with a series of spatial dataset including climate variability, ozone (O_3) pollution, nitrogen (N) deposition, land cover change, N fertilization, and elevated atmospheric carbon dioxide (CO_2), to examine the terrestrial fluxes of CH_4 and N_2O over the continental North America during 1979-2008. Over the study period, approximately 14.69 ± 1.64 T g C a^{-1} (1T g = 10^{12} g) of CH_4, with a 95% confidence interval of (5.95 T g C a^{-1}, 23.10 T g C a^{-1}), and 1.94 ± 0.16 T g N a^{-1} of N_2O, with a 95% confidence interval of (0.75 T g N a^{-1}, 3.38 T g N a^{-1}), were released from terrestrial ecosystems in North America. Both the United States and Canada acted as CH_4 sources to the atmosphere, yet Mexico mainly oxidized and consumed CH_4 from the atmosphere. Wetlands in North America contributed predominantly to the regional CH_4 source, while all other ecosystems acted as sinks for atmospheric CH_4, of which forests accounted for 36.8%. Regarding N_2O emission in North America, the United States, Canada, and Mexico contributed 56.19%, 18.23%, and 25.58%, respectively, to the continental source over the past 30 years. Forests and croplands were the two ecosystems that contributed most to the continental N_2O emission.

Our simulations indicate that over the past 30 years 440.75 ± 8.97 T g CH_4-C was released from North Americas' terrestrial ecosystems; baseline emission contributed 417.24 ± 6.83 T g CH_4-C and global change factors accumulatively contributed 23.51 ± 9.61 T g CH_4-C. O_3 pollution led to a reduced CH_4 emission by 2.30 ± 0.49 T g CH_4-C. All other factors including climate variability, N deposition, elevated atmospheric CO_2, N fertilizer application, and land conversion enhanced terrestrial CH_4 emissions by 19.80 ± 12.42, 0.09 ± 0.02, 6.80 ± 0.86, 0.01 ± 0.001, and 3.95 ± 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 ± 7.74 T g CH_4-C. From 1979 to 2008, North America's terrestrial ecosystems accumulatively emitted 58.17 ± 0.85 T g N_2O-N, of which global change factors contributed 2.81 ± 0.98 T g N_2O-N, and baseline emission contributed 55.35 ± 0.56 T g N_2O-N. The elevated CO_2 led to a decrease in terrestrial N_2O emission at 0.51 ± 0.07 T g N_2O-N. Climate variability, N deposition, O_3 pollution, N fertilization, and land use conversion increased N_2O emission by 0.56 ± 0.68, 0.50 ± 0.07, 0.10 ± 0.02, 0.92 ± 0.09, and 0.16 ± 0.01 T g N_2O-N, respectively. The interactive effect among multiple factors enhanced N_2O emission by 1.10 ± 0.37 T g N_2O-N over the 30 years. The factorial attribution of terrestrial fluxes of CH_4 and N_2O varied across countries.

Local sensitivity and uncertainty analyses indicate that the uncertainties in terrestrial CH_4 and N_2O fluxes varied across the continental North America; the largest uncertainty in CH_4 flux locates in wetland, while the largest uncertainty in N_2O flux lies in tropical forest, followed by cropland. This study provides useful and valuable information to both scientific community and policy makers such as magnitude, spatiotemporal and underlying mechanisms of terrestrial CH_4 and N_2O fluxes over the continental North America.

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List of Abbreviations

CERN	Chinese Ecological Research Network
CH_4	Methane
CO_2	Carbon dioxide
DLEM	Dynamic Land Ecosystem Model
DOC	Dissolved Organic Carbon
FAO	Food and Agriculture Organization
GPP	Gross Primary Productivity
HYDE	History Database of the Global Environment
LTER	Long-Term Ecological Research
NARR	North America Regional Reanalysis
N_2O	Nitrous oxide
NOAA	National Oceanic and Atmospheric Administration
NPP	Net Primary Productivity
O_3	Ozone
PFT	Plant Function Type
SOM	Soil Organic Matter
VEMAP	the Vegetation/Ecosystem Modeling and Analysis Project

Chapter 1. Introduction[1]

The global change has become a critical environmental problem faced by human society; it has altered and will be continuously altering the structure and function of terrestrial ecosystems on our planet in the 21st century. Influenced by climate system, the terrestrial ecosystems might change and feed back to climate system by releasing trace gases such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) etc (Denman et al., 2007). A better understanding of the interaction between the atmosphere and terrestrial ecosystems is of upmost importance for the global change research and the sustainability of human society (Denman et al., 2007; Schimel and Gulledge, 1998).

Terrestrial ecosystems could act as either sources or sinks for atmospheric CH_4 and N_2O, depending on the locale (Liu, 1996; Potter, 1997; Ridgwell et al., 1999; Chapuis-Lardy et al., 2007; Xu et al., 2008). Globally, natural sources from terrestrial ecosystems contribute approximately 40% to the atmospheric CH_4, and more than half to the atmospheric N_2O when removing oceanic contributions (Denman et al., 2007). Given that a relatively well-understood CO_2 flux has been achieved, quantifying the fluxes CH_4 and N_2O in the terrestrial ecosystems is critically important and urgent for further mitigation of climate change, especially at regional scale (Rigby et al., 2008; Forster et al., 2007; Potter, 1997; Ridgwell et al., 1999; Chapuis-Lardy et al., 2007; Xu et al., 2008).

[1] This dissertation uses the official format required by the journal *Biogeosciences* (www.biogeosciences.net)

North America, with its large land area and high proportion of natural wetland (approximately 30% of the global wetland) (Bridgham et al., 2006; Mitsch and Gosselink, 2007), plays a critical role in global carbon cycling (Schimel et al., 2000). However, only a few studies have simultaneously investigated CH_4 and N_2O fluxes over terrestrial ecosystems in North America (Bridgham et al., 2006).

Currently there are three general approaches to estimate trace gas fluxes: flux extrapolation method, inverse method, and process-oriented modeling approach (Liu 1996). Each method possesses its own advantages and shortcomings. Compared with flux extrapolation and inverse methods, the process-oriented modeling approach is based on the understanding of biogeochemical processes responsible for CH_4 and N_2O production and consumption; the regional application of process-oriented model usually provides reliable estimates since it considers the spatial heterogeneity of ecosystem attributes, namely soil, vegetation, climate, etc (Tian et al., 2010b).

Many factors can influence CH_4 and N_2O fluxes in terrestrial ecosystems at both site and regional levels, such as elevated atmospheric CO_2 (Hutchin et al., 1995; Schrope et al., 1999; Phillips et al., 2001b; Phillips et al., 2001a), tropospheric ozone (O_3) pollution (Morsky et al., 2008), nitrogen (N) input (Ding et al., 2004), climate change (Goldberg and Gebauer, 2009) and land cover change (Willison et al., 1995; Huang et al., 2010). However, most previous process-based modeling efforts did not take into account the concurrent effects of multiple global change factors (Potter, 1997; Cao et al., 1998; Walter et al., 2001; Zhuang et al., 2007; Zhuang et al., 2004). Large uncertainty still exists in the magnitudes, spatial and temporal patterns of CH_4 and N_2O fluxes at large scales (Kort et al., 2008; Christensen et al., 1996; Zhuang et al., 2004; Bridgham et al., 2006; Potter et al., 2006).

The uncertainties associated with modeling studies are another important issue for regional estimation of terrestrial fluxes of CH_4 and N_2O (Tian et al., 2010b; Tang & Zhuang, 2009). However, the uncertainties associated with the estimates are rarely investigated, especially at regional scale (Potter, 1997; Potter et al., 1996; Walter et al., 2001). This is due to both the complexity of uncertainty sources (Haefner, 2005), namely input data, model structure, and parameters, and the challenges in the approaches for uncertainty analysis (Tang and Zhuang, 2009; Varella et al., 2010; Werner et al., 2007). Bayesian Monte Carlo method has been widely adopted to estimate the uncertainties in the ecological studies (Knorr & Kattge, 2005; Lele et al., 2007). Thus, evaluation of the uncertainties associated with the simulated terrestrial fluxes of CH_4 and N_2O by using Bayesian Monte Carlo will be an important task in this study.

This study was conducted by enhancing and applying a process-based ecosystem model to address the following four interlinked questions:

1) **How much CH_4 and N_2O were released from or took up by the terrestrial ecosystems over the continental North America during 1979-2008?**
2) **How did the terrestrial CH_4 and N_2O flux vary across the continental North America at both temporal and spatial scales?**
3) **Which factor dominantly controlled the spatial and temporal variations in terrestrial CH_4 and N_2O fluxes?**
4) **How confident are we in the reported fluxes of CH_4 and N_2O over North America in terms of the uncertainties induced by parameterization process?**

To answer the four questions above, I conducted this study and addressed them one by one. In sequence, I first developed CH_4 and N_2O modules in the framework of the Dynamic Land Ecosystem Model (DLEM), and prepared the input data for model simulations; then the

model parameterization was conducted to determine the values of each parameter for model simulations; the simulations were set up by using the enhanced model and calibrated parameters, in conjunction with the consistent model driving data; the magnitude, spatial and temporal variations in terrestrial CH_4 and N_2O fluxes over the continental North America were examined; combining single factor simulations with all combined simulation, I attributed the spatiotemporal variations in terrestrial CH_4 and N_2O fluxes to multiple global change factors including climate variability, elevated atmospheric CO_2, N deposition, O_3 pollution, land conversion[2], and nitrogen fertilizer application; finally, using Bayesian Monte Carlo method, I estimated the potential uncertainties caused by parameterization process.

Therefore, this dissertation is formed under the following framework: chapter 1, introduces the basic scientific questions and significance of this study as examining the terrestrial fluxes of CH_4 and N_2O over the continental North America; chapter 2 presents the structure, parameterization and validation of CH_4 and N_2O modules in the framework of DLEM model, and further reports the magnitude of terrestrial CH_4 and N_2O fluxes as simulated by the DLEM model; chapter 3 introduces the further efforts to attribute the spatial and temporal variations in terrestrial CH_4 flux to multiple global change factors including climate variability, elevated atmospheric CO_2, N deposition, O_3 pollution, land cover conversion, and N fertilization; chapter 4 introduces the attribution of spatial and temporal variations in terrestrial N_2O flux to multiple global change factors; chapter 5 reports the method for uncertainty analysis, and the potential uncertainties induced by parameterization process; finally, chapter 6 provides a summary of the entire work, and concludes the major findings through this study.

[2] The land conversion in this study represents the conversion of land use type between natural vegetation and human disturbed land use types including cropland and urban area.

Chapter 2. Spatial and Temporal Patterns of CH_4 and N_2O Fluxes in Terrestrial Ecosystems of North America during 1979-2008: Application of a Global Biogeochemistry Model[3]

Abstract

Continental-scale estimations of terrestrial methane (CH_4) and nitrous oxide (N_2O) fluxes over a long time period are crucial to accurately assess the global balance of greenhouse gases and enhance our understanding and prediction of global climate change and terrestrial ecosystem feedbacks. Using a process-based global biogeochemical model, the Dynamic Land Ecosystem Model (DLEM), we quantified simultaneously CH_4 and N_2O fluxes in North America's terrestrial ecosystems from 1979 to 2008. During the past 30 years, approximately 14.69 ± 1.64 T g C a^{-1} (1T g = 10^{12} g) of CH_4, and 1.94 ± 0.16 T g N a^{-1} of N_2O were released from terrestrial ecosystems in North America. At the country level, both the United States and Canada acted as CH_4 sources to the atmosphere, but Mexico mainly oxidized and consumed CH_4 from the atmosphere. Wetlands in North America contributed predominantly to the regional CH_4 source, while all other ecosystems acted as sinks for atmospheric CH_4, of which forests accounted for 36.8%. Regarding N_2O emission in North America, the United States, Canada, and Mexico contributed 56.19%, 18.23%, and 25.58%, respectively, to the continental source over the past 30 years. Forests and croplands were the two ecosystems that contributed most to continental N_2O

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emission. The inter-annual variations of CH_4 and N_2O fluxes in North America were mainly attributed to year-to-year climatic variability. While only annual precipitation was found to have a significant effect on annual CH_4 flux, both mean annual temperature and annual precipitation were significantly correlated to annual N_2O flux. The regional estimates and spatiotemporal patterns of terrestrial ecosystem CH_4 and N_2O fluxes in North America generated in this study provide useful information for global change research and policy making.

1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are two potent greenhouse gases which in sum contribute to more than one fourth of global warming caused by anthropogenic activities (Forster et al., 2007). Although the concentrations of CH_4 and N_2O in the atmosphere are relatively low, their warming potentials are much higher than that of carbon dioxide (Denman et al., 2007). CH_4 and N_2O also play significant roles in ozone layer chemistry (Denman et al., 2007; Forster et al., 2007). Similar to the increase of atmospheric CO_2 concentration, the concentrations of these two gases dramatically increased since the Industrial Revolution (Forster et al., 2007; Tueut et al., 2007; Rigby et al., 2008). Although the importance of CH_4 and N_2O emissions in changing the Earth's climate has been recognized, scientific community has placed large emphasis on the CO_2 problem. Understanding and quantifying CH_4 and N_2O fluxes in terrestrial ecosystems at large spatial scales, therefore, becomes an urgent task for accurately predicting the future climate change (Rigby et al., 2008; Forster et al., 2007; Sheldon and Barnhart, 2009).

Terrestrial ecosystems could act as either sources or sinks for atmospheric CH_4 and N_2O, depending on the time and location (Liu, 1996; Potter, 1997; Ridgwell et al., 1999; Chapuis-Lardy et al., 2007; Xu et al., 2008). Globally, natural sources from terrestrial ecosystems

contribute approximately 40% to the CH_4, and more than half to the N_2O releases to the atmosphere when removing oceanic contribution (Denman et al., 2007). North America, with its large land area and high proportion of natural wetland (approximately 30% of the global wetland) (Bridgham et al., 2006; Mitsch and Gosselink, 2007), plays a critical role in global carbon cycling (Schimel et al., 2000). However, only a few studies have investigated CH_4 and N_2O fluxes over terrestrial ecosystems in North America (Bridgham et al., 2006). For example, Zhuang et al (2004) estimated that soils in Canada and Alaska emitted 7.1 and 3.8 T g CH_4 a^{-1}, respectively, during the 1990s. Bridgham et al (2006) estimated that CH_4 emission in North America's wetlands is 9 T g CH_4 a^{-1}. Using a satellite-derived modeling approach, Potter et al. (2006) estimated that the CH_4 emission from the natural wetlands in the conterminous United States is 5.5 T g CH_4 a^{-1}. Several studies also reported the fluxes of N_2O in terrestrial ecosystems at global and regional scales using empirical approaches (Xu et al., 2008). While these studies improved our understanding of CH_4 and N_2O fluxes in North America, accurate estimations of terrestrial ecosystem CH_4 and N_2O fluxes in the entire continent over a long time period are still needed (Wofsy and Harriss, 2002).

Many factors can influence CH_4 and N_2O fluxes in terrestrial ecosystems at site and regional levels, such as elevated CO_2 (Hutchin et al., 1995; Schrope et al., 1999; Phillips et al., 2001b; Phillips et al., 2001a), tropospheric ozone pollution (Morsky et al., 2008), nitrogen input (Ding et al., 2004), climate change (Goldberg and Gebauer, 2009) and land cover change (Willison et al., 1995; Huang et al., 2010). However, most previous process-based modeling efforts did not take into account the concurrent effects of multiple global change factors (Potter, 1997; Cao et al., 1998; Walter et al., 2001; Zhuang et al., 2007; Zhuang et al., 2004). Large uncertainty still exists in the magnitudes, spatial and temporal patterns of CH_4 and N_2O fluxes at

large scales (Kort et al., 2008; Christensen et al., 1996; Zhuang et al., 2004; Bridgham et al., 2006; Potter et al., 2006).

Recently, we developed a process-based biogeochemistry model, the Dynamic Land Ecosystem Model (DLEM), to simulate biogeochemical cycling of carbon, nitrogen and water in the land ecosystems. The DLEM considers multiple factors including climate, atmospheric compositions (CO_2, O_3), precipitation chemistry (nitrogen composition), natural disturbances (fire, insect/disease, hurricane, etc), land-use/land-cover change, and land management (harvest, rotation, fertilization, irrigation, etc) (Tian et al., 2005, 2008, 2010a, 2010b; Ren et al., 2007a, 2007b, 2009; Zhang et al., 2007, 2008; Lu, 2009; Liu et al., 2008; Chen et al., 2006; Xu, 2010). This model has been successfully applied to simulate the effects of multiple environmental factors on carbon and water cycles in China (Ren et al., 2007a, 2007b, Lu, 2009; Liu et al., 2008; Chen et al., 2006, Xu, 2010) and USA (Tian et al., 2008, 2010a, 2010b; Zhang et al., 2007, 2008).

In this study, we enhanced the model's capability by addressing the biogeochemical processes of CH_4 and N_2O and simulated CH_4 and N_2O fluxes over terrestrial ecosystems in North America from 1979 to 2008. The objectives of this study are: 1) to develop the CH_4 and N_2O modules in the framework of an extant process-based model, DLEM; 2) to compare modeled results with field observations and other regional estimates; 3) to estimate CH_4 and N_2O fluxes in North America's terrestrial ecosystems from 1979 to 2008; 4) to quantify the contributions of individual countries and biomes to regional CH_4 and N_2O fluxes in North America.

2. Methodology

2.1. The DLEM model and its trace gas modules

The Dynamic Land Ecosystem Model (DLEM) couples major biogeochemical cycles, hydrological cycles, and vegetation dynamics to make daily, spatially-explicit estimates of carbon, nitrogen, and water fluxes and pool sizes (C and N) in terrestrial ecosystems. There are five core components in the DLEM: 1) biophysics, 2) plant physiology, 3) soil biogeochemistry, 4) dynamic vegetation, and 5) disturbance, land use and management. Briefly, the biophysics component simulates the instantaneous fluxes of energy, water, and momentum within land ecosystems and their exchanges with the surrounding environment. The plant physiology component simulates major physiological processes, such as plant phenology, C and N assimilation, respiration, allocation, and turnover. The soil biogeochemistry component simulates the dynamics of nutrient compositions and major microbial processes. The biogeochemical processes, including the nutrient mineralization/immobilization, nitrification/denitrification, decomposition, and methane production/oxidation are considered in this component. The dynamic vegetation component simulates the structural dynamics of vegetation caused by natural and human disturbances. Two processes are considered: the biogeography redistribution when climate change occurs, and the recovery and succession of vegetation after disturbances. Like most dynamic global vegetation models, the DLEM builds on the concept of plant functional types (PFT) to describe vegetation attributes. The disturbances, land use and management component simulates cropland conversion, reforestation after cropland abandonment, and forest management practices such as harvest, thinning, fertilization and prescribed fires.

The interactions and feedbacks of various processes among core components are simulated as controls or material flows (Fig 1). The biophysics component yields influences on plant physiology component through the effects of water, temperature and radiation, and on soil biogeochemistry component through the effects of soil moisture and temperature; the plant physiology component yields influences on the biophysics component through changes in leaf area index (LAI), canopy conductance, and transpiration, on the soil biogeochemistry component through litter-fall, and on the dynamic vegetation component through biomass growth; the dynamic vegetation component yields influences on the plant physiology and soil biogeochemistry components through shifts of plant function type (PFT); the soil biogeochemistry component yields influences on the dynamics vegetation and plant physiology components through nutrient flow; disturbances, land use and management component yields influences on the other four components through changes in land cover type, PFT and nutrient and water flow (Fig 1).

Meanwhile, the DLEM uses climate data from regional climate and atmosphere chemistry component which could be a climate model or input data. The DLEM outputs including ecosystem carbon and nitrogen pools and fluxes (e.g. greenhouse gases) will enter the atmosphere; and the water output and associated nutrients from the DLEM will enter water transport module and flow into lake, river and ocean. All the components are also linked together by water and energy fluxes (Fig 1).

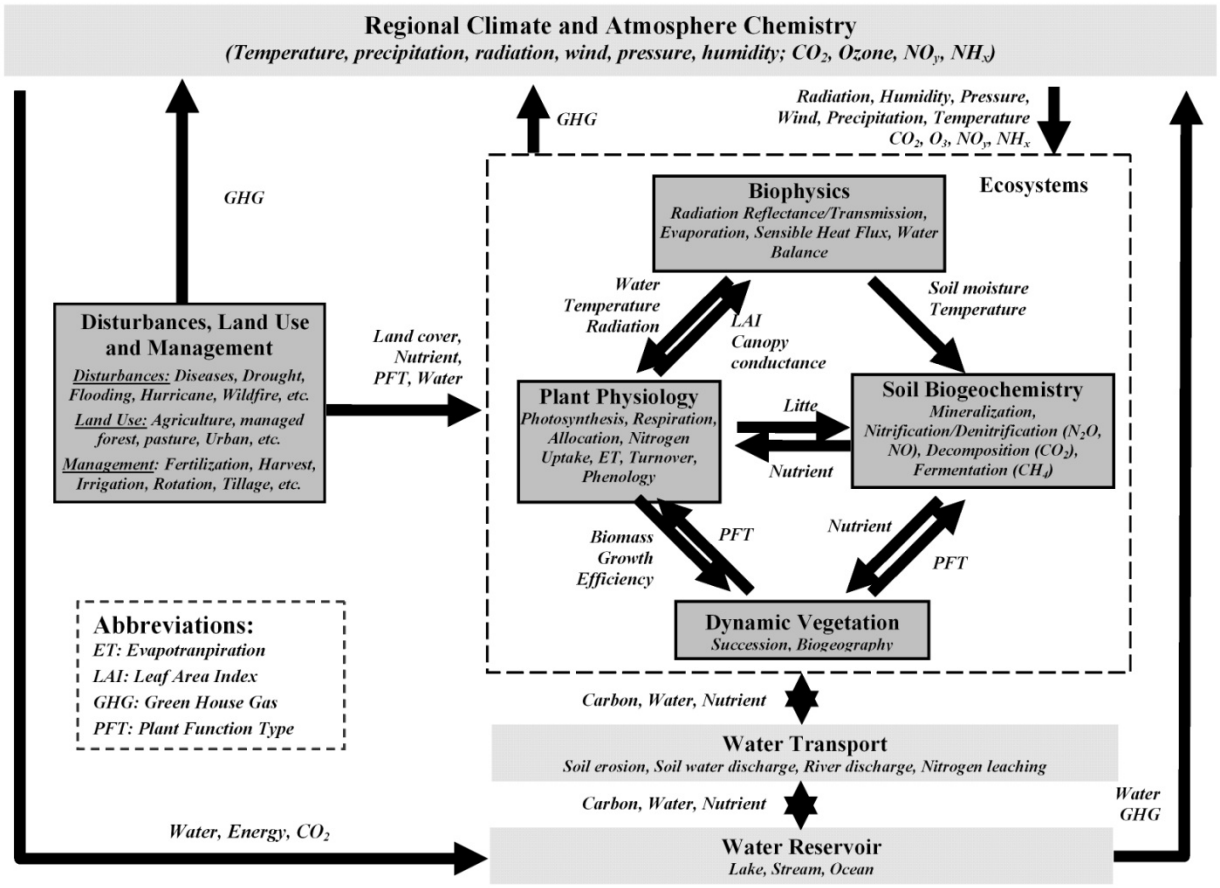
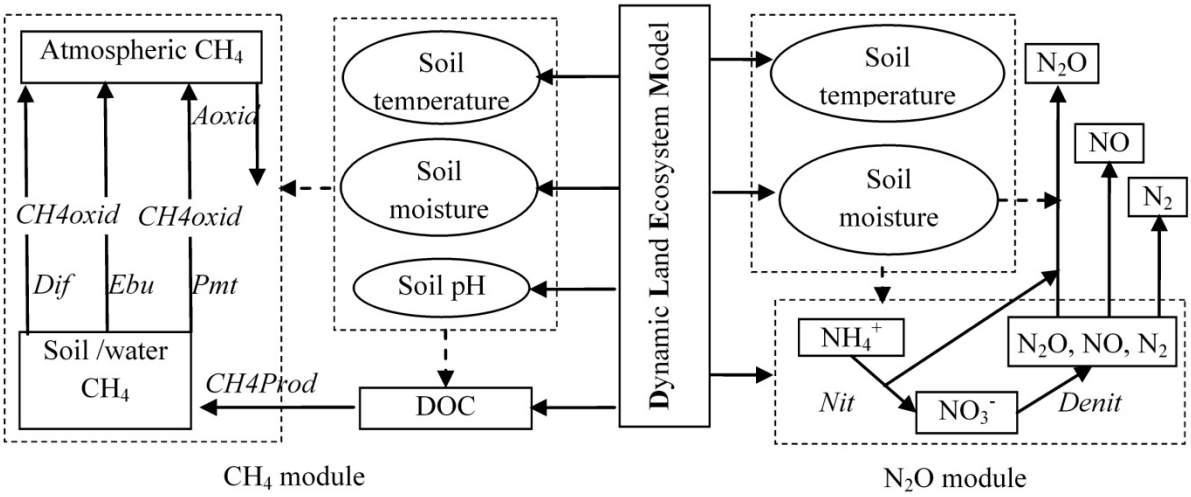


Fig. 1 Conceptual model of the Dynamic Land Ecosystem Model (DLEM) (Five core components are included in the DLEM)

The DLEM emphasizes the modeling and simulation of managed ecosystems including agricultural ecosystems, plantation forests and pastures. The spatial data sets of land management, such as irrigation, fertilization, 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 US Long-Term Ecological Research (LTER) network, AmeriFlux network, and the Chinese Ecological Research Network (CERN) which cover various ecosystems, including forests, grasslands, shrub, tundra, desert, wetland, and croplands. The major carbon, nitrogen and water variables have been validated with observational data. The simulated results have been compared with independent

field data and satellite products. The DLEM operates at a daily time step and at varied spatial resolutions, from meters to kilometers, from regional to global. The additional information on the processes, interactions and feedbacks in the DLEM and associated input/output data (Fig. 1) can be found in our previous studies (Tian et al., 2005, 2008, 2010a, 2010b; Ren et al., 2007a, 2007b, 2009; Zhang et al., 2007, 2008; Lu, 2009; Liu et al., 2008; Chen et al., 2006; Xu et al., 2010).

In this paper, we provide a detailed description of the CH_4 and N_2O modules with an emphasis on major processes that control fluxes of CH_4 and N_2O in terrestrial ecosystems (Fig. 2).



Major processes: *Aoxid*: Atmospheric CH_4 oxidation; *CH4prod*: CH_4 production; *CH4oxid*: CH_4 Oxidation during diffusion and ebullition transport; *CH4oxidp*: CH4 oxidation during plant-mediated transport (Occur in herbaceous wetland only); *Dif*: CH_4 diffusion transport; *Ebu*: CH_4 ebullition transport; *Pmt*: Plant-mediated transport of CH_4; (Occur in herbaceous wetland only); *Nit*: Nitrification; *Denit*: Denitrification;

DLEM provides the environment factors and substrate for CH_4 and N_2O modules; the environmental controls were shown as dash lines.

Fig. 2 Modules of CH_4 and N_2O in the Dynamic Land Ecosystem Model (DLEM) (CH_4 production, oxidation, and transport are considered in the CH_4 module; nitrification and nitrogen reduction are considered in the N_2O module)

2.1.1. The CH_4 module

The CH_4 exchanges between ecosystems and the atmosphere are a combination of CH_4 production, oxidation, and transportation from soil pore water to the atmosphere. The DLEM only considers CH_4 production from dissolved organic carbon (DOC), which is indirectly controlled by environmental factors including soil pH, temperature and soil moisture content. The production of DOC mainly comes from two sources: allocation of gross primary production (GPP) and decomposition of litter-fall and soil organic matter. The accumulated DOC is either used as substrate for methane, leaves system as leaching, or enters the atmosphere as CO_2 via decomposition. CH_4 oxidation, including the oxidation during CH_4 transport to the atmosphere, CH_4 oxidation in the soil pore water, and atmospheric CH_4 oxidation on the soil surface, is determined by CH_4 concentrations in the air or soil pore water, as well as soil moisture, pH, and temperature. Most CH_4-related biogeochemical reactions in the DLEM are described by using the Michaelis-Menten equation with two coefficients: maximum reaction rate and half saturation coefficient. Three pathways for CH_4 transport from soil to the atmosphere include ebullition, diffusion, and plant-mediated transport. It is assumed that methane-related biogeochemical processes only occur in the top 50 cm of soil profile. The net CH_4 flux between the atmosphere and soil is determined by the following equation:

$$\boldsymbol{F_{CH4} = F_P + F_D + F_E - F_{air,oxid} - F_{trans,oxid}} \qquad \text{Equation 1}$$

where F_{CH4} is the flux of CH_4 between soil and the atmosphere (g C m^{-2} d^{-1}); F_P is plant-mediated transport from soil pore water to the atmosphere (g C m^{-2} d^{-1}); F_D is the diffusive flux of CH_4 from water surface to the atmosphere (g C m^{-2} d^{-1}); F_E is the ebullitive CH_4 emission to the atmosphere; $F_{air,\ oxid}$ is the rate of atmospheric methane oxidation (g C m^{-2} d^{-1}); $F_{trans,\ oxid}$ is the oxidized CH_4 during plant-mediated transport (g C m^{-2} d^{-1}).

The concentration of CH_4 in the soil pore water was governed by the following equations:

$$\frac{d[CH4]}{dt} = f([CH4]) = CH4_{prod} - \frac{F_P}{H} - \frac{F_D}{H} - \frac{F_E}{H} - CH4_{soil,oxid} \quad \text{Equation 2}$$

where $[CH_4]$ is the concentration of CH_4 in water (g C m^{-3}); $CH4_{prod}$ is the production of CH_4 in soil pore water (g C m^{-3} d^{-1}); $CH4_{soil,\ oxid}$ is the oxidation rate of CH_4 in soil pore water (g C m^{-3} d^{-1}); H is the soil depth of the first layer for methane production and oxidation.

2.1.1.1. CH_4 production

The production of CH_4 in soil pore water is controlled by the concentration of DOC and environmental factors (Eq. 2),

$$CH4_{prod} = V_{prod,max} \times \frac{[DOC]}{[DOC]+Km_{prod}} \times f(T_{soil}) \times f(pH) \times f_{prod}(vwc) \quad \text{Equation 3}$$

where $V_{prod,\ max}$ is the maximum rate of CH_4 production (g C m^{-3} d^{-1}), $[DOC]$ is the concentration of DOC (g C m^{-3}); Km_{prod} is the half-saturation coefficient of CH_4 production (g C m^{-3}); $f(T_{soil})$ is a multiplier that describes the effect of soil temperature on CH_4 production and oxidation; $f(pH)$ is a multiplier that describes the effect of soil pH on CH_4 production and oxidation; $f_{prod}(vwc)$ is a multiplier that describes the effect of soil moisture on CH_4 production.

2.1.1.2. CH_4 oxidation

Three pathways are considered in the DLEM for CH_4 oxidation: 1) atmospheric CH_4 oxidation, also called the diffusion processes of CH_4 from the atmosphere to the soil pore water, mainly simulates the oxidation of atmospheric CH_4 in the soil pore water; 2) the process of CH_4 oxidation in the soil pore water, mainly simulates the oxidation of CH_4 which is dissolved in water or accumulated in soil porosity; and 3) the process of CH_4 oxidation that occurs during the plant-mediated transport of CH_4 from soil pore water to the atmosphere. The DLEM assumes that the process of CH_4 oxidation in soil pore water includes the CH_4 oxidation during ebullition and diffusion because these two processes only occur in water.

2.1.1.2.1. Atmospheric CH_4 oxidation

Oxidation of atmospheric CH_4 is estimated as:

$$\boldsymbol{F_{air,oxid} = V_{air,oxid,max} \times \frac{[AtmCH4]}{[AtmCH4]+Km_{air,oxid}} \times f(T_{soil}) \times f(pH) \times f_{oxid}(vwc)} \quad \text{Equation 4}$$

where $V_{air, oxid, max}$ is the maximum oxidation rate of atmospheric CH_4 (g C m^{-2} d^{-1}); , $km_{air, oxid}$ is the half saturation coefficient of atmospheric CH_4 oxidation (g C m^{-3}); [*AtmCH4*] is the atmospheric CH_4 concentration (g C m^{-3}); $f_{oxid}(vwc)$ is a multiplier that describes the effect of soil moisture on atmospheric CH_4 oxidation. Because the atmospheric CH_4 oxidation is mainly carried out by soil methanotrophy, and low soil organic matter means lower soil microbial biomass (Conrad, 1996), the DLEM assumes that there is no atmospheric CH_4 oxidation when soil organic matter is less than 10 g C m^{-2}.

2.1.1.2.2. CH_4 oxidation during plant-mediated transport

During the process of plant-mediated CH_4 transport from soil to the atmosphere, portions of CH_4 will be oxidized at the rate of:

$$\boldsymbol{F_{trans,oxid} = \min\left(V_{trans,oxid,max} \times \frac{F_P}{F_P+Km_{trans,oxid}} \times f(T_{air}), F_P\right)} \quad \text{Equation 5}$$

where $F_{trans, oxid}$ is the oxidation rate of CH_4 during plant-mediated transport (g C m^{-2} d^{-1}); $V_{trans, oxid, max}$ is the maximum rate of CH_4 oxidation (g C m^{-2} d^{-1}); $Km_{trans, oxid}$ is the half saturation coefficient of soil CH_4 oxidation during transportation (g C m^{-2}); T_{air} is the air temperature; $f(T_{air})$ is a multiplier that represents the effect of air temperature on the oxidation of CH_4 during plant-mediated transport.

2.1.1.2.3. Soil pore water CH_4 oxidation

The accumulated CH_4 in soil pore water is oxidized at the rate of:

$$\boldsymbol{CH4_{soil,oxid}} = \mathbf{min}\left(\boldsymbol{V_{soil,oxid,max}} \times \frac{[\boldsymbol{CH4}]}{[\boldsymbol{CH4}]+\boldsymbol{Km_{soil,oxid}}} \times \boldsymbol{f}(\boldsymbol{T_{soil}}) \times \boldsymbol{f}(\boldsymbol{pH}) \times \boldsymbol{f_{oxid}}(\boldsymbol{vwc}), [\boldsymbol{CH4}]\right)$$

Equation 6

where $V_{soil, oxid, max}$ and $Km_{soil, oxid}$ are maximum soil pore water CH_4 oxidation rate (g C m^{-3} d^{-1}) and half saturation coefficient of CH_4 oxidation in soil pore water (g C m^{-3}), respectively; *[CH4]* is the concentration of CH_4 in soil pore water (g C m^{-3}).

2.1.1.3. CH_4 transport

In this model, ebullition, diffusion and plant-mediated transport, are considered the three pathways by which CH_4 can be transported from soil pore water to the atmosphere.

2.1.1.3.1. Ebullition

The ebullition transport of CH_4 from water to the atmosphere is estimated as:

$$\boldsymbol{F_E} = \mathbf{max}\left(([\boldsymbol{CH4}] - \mathbf{6}), \mathbf{0}\right) \times \boldsymbol{H} \quad \text{Equation 7}$$

where F_E is the flux of CH_4 from water to the atmosphere via ebullition (g C m^{-2} d^{-1}); *6* is the threshold value above which the dissolved CH_4 will form bubbles and leave water (g C m^{-3}), and is equals to 0.5 mol CH_4 m^{-3} (Walter et al., 2001). Because this process occurs in very short time (Walter et al., 2001;Zhuang et al., 2004), the DLEM assumes that all the dissolved CH_4 above this threshold value will leave water via bubbles in one day.

2.1.1.3.2. Plant-mediated transport

The plant-mediated CH_4 emission from water to the atmosphere is estimated as:

$$\boldsymbol{F_P} = \boldsymbol{V_{plant,trans}} \times ([\boldsymbol{CH4}] - [\boldsymbol{CH4}]_{\boldsymbol{max}}) \times \mathbf{min}\left(\frac{\boldsymbol{GPP}}{\boldsymbol{GPP_{max}}}, \mathbf{1}\right) \quad \text{Equation 8}$$

$$[\boldsymbol{CH4}]_{\boldsymbol{max}} = [\boldsymbol{AtmCH4}] \times \boldsymbol{\beta} \quad \text{Equation 9}$$

where F_P is the CH_4 transport via vascular plant (g C m^{-2} d^{-1}); $V_{plant, trans}$ is the transport coefficient of CH_4 transportation through plant (m d^{-1}), which is set as 0.68 (Kettunen, 2003);

$[CH4]_{max}$ is the maximum CH_4 concentration in soil solution (g C m^{-3}); *GPP* is the gross primary productivity of current day (g C m^{-2} d^{-1}); GPP_{max} is the maximum daily GPP (g C m^{-2} d^{-1}), which is set as 5 in this study; β is the Bunsen solubility coefficient (0.035 ml ml^{-1}) (Yamamoto et al., 1976). Since there is no report on the plant-mediated transport of CH_4 by woody plant, the DLEM assumes that the plant-mediated transport only occurs in herbaceous biomes; F_P is set to zero for all woody ecosystems.

2.1.1.3.3. Diffusion

The DLEM treats the top 0.5 m of the soil profile as one layer, and the CH_4 generated under water's surface is assumed to have a fast diffusion rate to water's surface. The diffusion estimated here is the exchange of CH_4 between the water surface and the atmosphere.

$$\boldsymbol{F_D = V_{exchange} \times ([CH4] - [CH4]_{max})} \quad \text{Equation 10}$$

where $V_{exhange}$ is the exchange coefficient of CH_4 through the interface of soil pore water and the atmosphere (m d^{-1}); it is set as 0.3 m d^{-1} (Happell and Chanton, 1995).

2.1.1.4. Environmental factors affecting methane processes

To simulate the environmental effects on methane production, oxidation and transport, the DLEM considers three environmental factors: soil pH, soil moisture, and temperature. These three factors have been considered as the most important external factors on CH_4 production, consumption, and transport (Cao et al., 1995; Huang et al., 1998; Mer and Roger, 2001; Zhuang et al., 2004). The line graphs showing the environmental controls on CH_4 production and consumption could be found in Fig. 3.

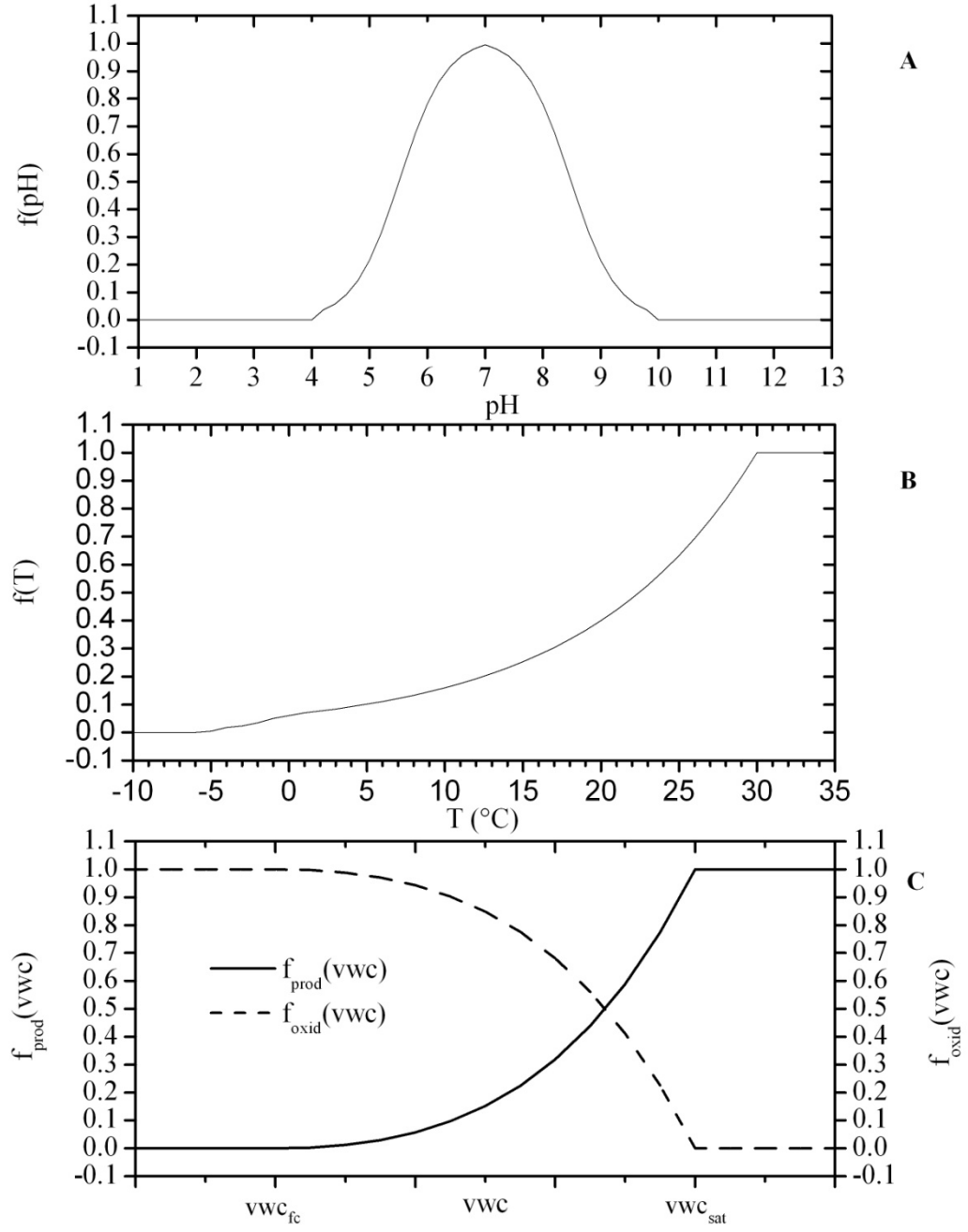


Fig. 3 Graphs showing environmental effects on methane production and oxidation (A: pH effects; B: Temperature effect; C: Moisture effects on methane production and oxidation)

In the DLEM, the effect of soil pH on methane production and oxidation (*f(pH)*) is calculated as a bell shape curve, following Cao et al. (1995) and Zhuang et al. (2004). Given that a number of reports showing CH_4 production and consumption at the circumstances of pH < 5 or

pH > 9 (Amaral et al., 1998; Mer and Roger, 2001; Sorokin et al., 2000), we set the effects of soil pH on CH_4 production and oxidation to zero when soil pH is smaller than 4 or larger than 10 (Eq. 11), which is different from Zhuang et al (2004) and Cao et al (1995).

$$f(pH) = \begin{cases} 0 & pH \le 4.0 \text{ or } pH \ge 10.0 \\ \frac{1.02}{1+1000000\times e^{(-2.5\times pH)}} & 4.0 < pH < 7.0 \\ \frac{1.02}{1+1000000\times e^{(-2.5\times(14.0-pH))}} & 7.0 < pH < 10.0 \end{cases} \quad \text{Equation 11}$$

where pH is the pH value of the soil profile.

The effect of temperature on methane processes (*f(T)*) is estimated by Q_{10} response curve which has been used by Huang et al (1998). The difference between our model and Huang et al's (1998) model is that we set the Q_{10} as 2.5, rather than 3.

$$f(T) = \begin{cases} 0.0 & T < -5 \\ Q_{10}^{\frac{T-30}{10}} & 30 > T \ge -5 \\ 1 & T \ge 30 \end{cases} \quad \text{Equation 12}$$

where Q_{10} is a scalar for the temperature sensitivity; T is temperature of soil or air.

The effect of soil moisture on methane processes is estimated based on the volumetric water content in the top soil layer (50 cm). Methane production and methane oxidation have reciprocal responsive curves to soil moisture.

$$f_{prod}(vwc) = \begin{cases} 0 & vwc \le vwc_{fc} \\ (\frac{vwc-vwc_{fc}}{vwc_{sat}-vwc_{fc}})^2 \times 0.368 \times e^{(\frac{vwc-vwc_{fc}}{vwc_{sat}-vwc_{fc}})} & vwc_{fc} < vwc < vwc_{sat} \\ 1 & vwc \ge vwc_{sat} \end{cases}$$

Equation 13

$$f_{oxid}(vwc) = 1 - f_{prod}(vwc) \quad \text{Equation 14}$$

where *vwc* is the volumetric water content of the top soil layer; vwc_{fc} is the field capacity and vwc_{sat} is the saturated water content. It is assumed that when the soil water content of an upland ecosystem is greater than field capacity, portions of the extra water will percolate or leave the system as base-flow.

2.1.2. The N_2O module

In the DLEM, both denitrification and nitrification processes are simulated as one-step process because we do not consider the mid-products in each process.

2.1.2.1. Nitrification

Nitrification, a process converting ammonium into nitrate, is simulated as a function of soil temperature, moisture, and soil NH_4^+ concentration (Lin et al., 2000).

$$\boldsymbol{N_{nit} = \min\left(t \times N_{pot,nit}, N_{NH4}\right)} \quad \text{Equation 15}$$

$$\boldsymbol{N_{pot,nit} = V_{nit,max} \times \frac{N_{NH4}}{N_{NH4}+Km_{nit}} \times f_{nit}(T_{soil}) \times f_{nit}(vwc)} \quad \text{Equation 16}$$

$$\boldsymbol{f_{nit}(T_{soil}) = Q_{10,nit}^{\left(\frac{T_{soil}-T_{opt,nit}}{10}\right)}} \quad \text{Equation 17}$$

$$\boldsymbol{f_{nit}(vwc) = \begin{cases} 1.17 \times \frac{vwc}{vwc_{fc}} + 0.165 & vwc < vwc_{fc} \\ 1 - 0.1 \times \frac{vwc}{vwc_{fc}} & vwc \geq vwc_{fc} \end{cases}} \quad \text{Equation 18}$$

where N_{nit} is the nitrification rate (g N m^{-3} d^{-1}); t is the time step as one day in this study; $N_{pot,\,nit}$ is the potential nitrification rate (g N m^{-3} d^{-1}); N_{NH4} is the concentration of NH_4^+ in the soil (g N m^{-3}); $V_{nit,\,max}$ is a parameter describing potential nitrification rate without limitation (g N m^{-3} d^{-1}); Km_{nit} is the half-saturation concentration of soil NH_4^+ for the maximum nitrification rate (g N m^{-3}); $f_{nit}(T_{soil})$ is a multiplier that describes the effect of soil temperature on nitrification; T_{soil} is the soil temperature (°C); $f_{nit}(vwc)$ is a multiplier that describes the effect of water content on nitrification (Lin et al., 2000; Riedo et al., 1998); $Q_{10,nit}$ is the temperature sensitivity of nitrification, which is set as 2; $T_{opt,nit}$ is the optimum temperature for nitrification, which is set as 20°C following Rideo et al. (1998) and Lin et al (2000); vwc is the volumetric water content; and vwc_{fc} is the soil field capacity.

2.1.2.2. Denitrification

Denitrification, through which the nitrate is converted into nitrogen gas, is simulated in the DLEM as a function of soil temperature, moisture, and soil NO_3^- concentration (Lin et al., 2000).

$$\boldsymbol{N_{denit} = \min\left(t \times N_{pot,denit}, N_{NO3}\right)} \quad \text{Equation 19}$$

$$\boldsymbol{N_{pot,denit} = V_{denit,max} \times \frac{N_{NO3}}{N_{NO3}+Km_{denit}} \times f_{denit}(T_{soil}) \times f_{denit}(vwc)} \quad \text{Equation 20}$$

$$\boldsymbol{f_{denit}(T_{soil}) = Q_{10,denit}^{\frac{T_{soil}-T_{opt,denit}}{10}}} \quad \text{Equation 21}$$

$$\boldsymbol{f_{denit}(vwc) = \begin{cases} 0.0 & vwc < vwc_{fc} \\ \frac{vwc}{vwc_{fc}} & vwc \geq vwc_{fc} \end{cases}} \quad \text{Equation 22}$$

where N_{denit} is the denitrification rate (g N m^{-3} d^{-1}); $N_{pot,\ denit}$ is the potential denitrification rate (g N m^{-3} d^{-1}); N_{NO3} is the concentration of NO_3^- in the soil (g N m^{-3}); $V_{denit,\ max}$ is a parameter describing potential denitrification rate without limitation (g N m^{-3} d^{-1}); Km_{denit} is the half-saturation concentration of soil NO_3^- for the maximum denitrification rate (g N m^{-3}); $f_{denit}(T_{soil})$ is a multiplier that describes the effect of soil temperature on denitrification; $f_{denit}(vwc)$ is a multiplier that describes the effect of water content on denitrification (Lin et al., 2000; Riedo et al., 1998); $Q_{10,denit}$ is the temperature sensitivity of denitrification, which is set as 3; and $T_{opt,denit}$ is the optimum temperature for denitrification, which is set as 25°C following Lin et al (2000).

2.1.2.3. N_2O emission

All products of denitrification and portion products of nitrification are nitrogen-containing gases. The empirical equation reported by Davidson et al (2000) is used to separate N_2O from other gases (mainly NO and N_2).

$$\boldsymbol{F_{N2O} = (0.001 * N_{nitrif+}N_{denitrif}) \times \frac{10^{vwc/\emptyset \times 0.026-1.66}}{(1+10^{vwc/\emptyset \times 0.026-1.66})}} \quad \text{Equation 23}$$

where F_{N2O} is the fluxes of N_2O from soil to the atmosphere (g N m^{-3} d^{-1}), 0.001 is the proportion of nitrification product released as gaseous nitrogen (Lin et al., 2000), and it is converted to fluxes in the unit area (g N m^{-2} d^{-1}) by multiplying the depth of the first soil layer (0.5m); $\emptyset$ is the soil porosity.

2.2. Input data preparation, model initialization and simulation

We developed gridded (32km × 32km), geo-referenced, time-series input data sets of climate (including daily temperature, precipitation, humidity, and solar radiation), annual nitrogen deposition rate, annual land-cover change and land management practices (including fertilization, irrigation) for the entire North America (including Canada, the United States, and Mexico). The climate dataset was generated based on North American Regional Reanalysis (NARR) dataset (http://nomads.ncdc.noaa.gov/data.php?name=access#narr_datasets). The maximum, minimum and average temperatures were calculated based on the eight 3-hour average 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 developed by History Database of the Global Environment (HYDE 3). Ozone data was retrieved from the global dataset developed by Felzer et al. (2005) covering 1900-2050. Annual nitrogen deposition data were retrieved from a global data set that was extrapolated from a three yearly maps (Dentener et al., 2006). Soil properties data, including soil texture, soil pH, soil bulk density, were extracted from a global data set Global Soil Data Task posted online on the Oak Ridge National Laboratory (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 source (www.cfi.ca). All the datasets were transformed and re-projected to

a consistent projection system for driving the DLEM model. The annual atmospheric concentration of CO_2 before 1959 was estimated by VEMAP (The Vegetation/Ecosystem Modeling and Analysis Project), and the data after 1959 were provided by National Oceanic and Atmospheric Administration (NOAA) (www.esrl.noaa.gov). The distributional map of contemporary vegetation types (Fig. 4) was developed using different sources of data, including global land-cover derived from Landsat imagery (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). It is assumed that the ice-covered land is not capable of producing or taking up CH_4 and N_2O, and thus was not considered in present simulation.

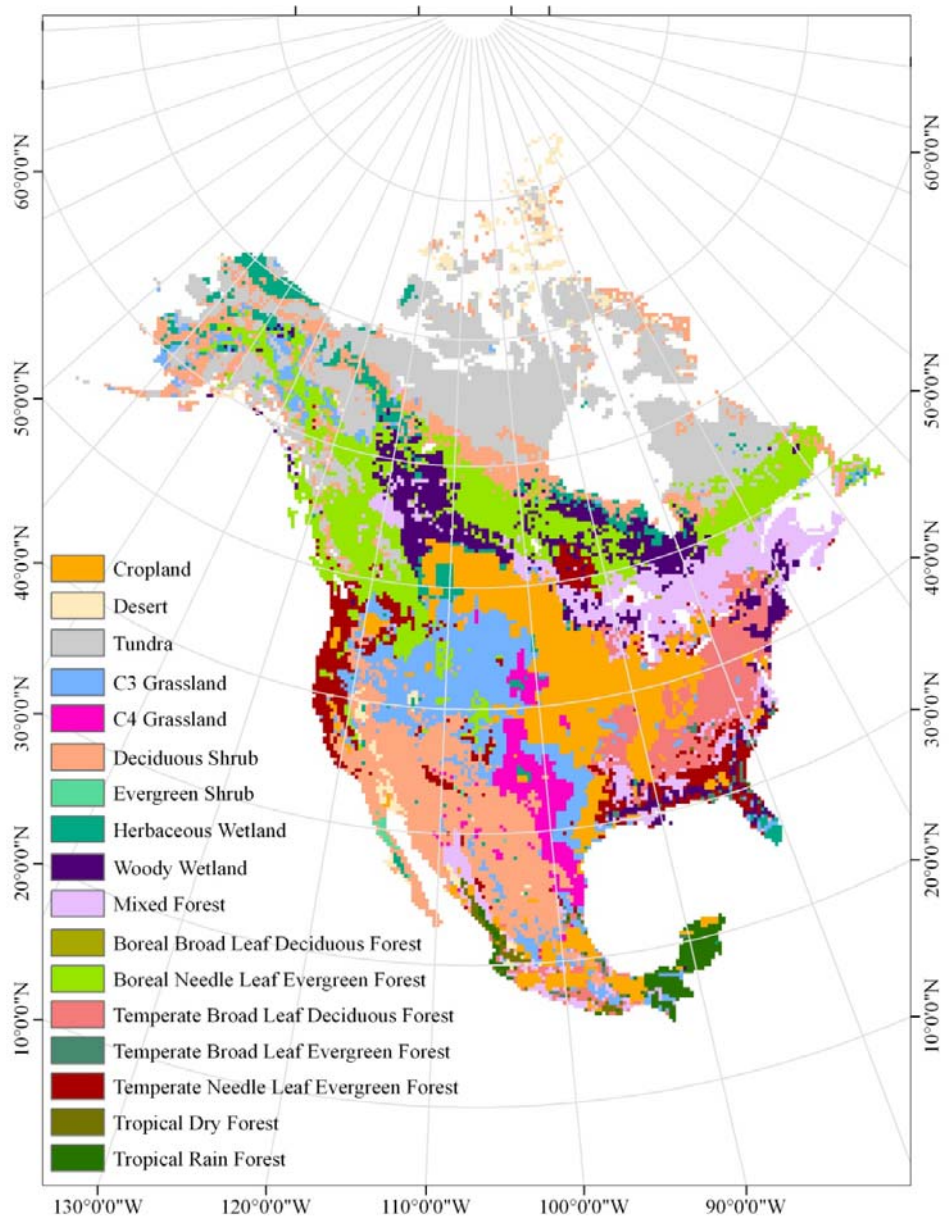


Fig. 4 Contemporary vegetation map used in this study (The year 2000 is shown)

The implementation of DLEM simulation includes the following runs: 1) equilibrium run, 2) spinning-up run and 3) transient run. In this study, we used potential vegetation, long-term mean climate during 1979-2008, the concentration levels of nitrogen deposition, ozone, atmospheric CO_2 in the year 1900 to drive the model run to an equilibrium state (i.e. the inter-

annual variations are < 1 g m^{-2} for carbon storage, < 0.1 g m^{-2} for nitrogen 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 nitrogen deposition inputs from 1901 to 2008 to simulate CH_4 and N_2O fluxes. The annual climate data between 1901 and 1978 were developed by randomly assigning a year between 1979 and 2008. Only the outputs between 1979 and 2008 were analyzed to show the spatial and temporal patterns of CH_4 and N_2O fluxes in North America's terrestrial ecosystems. Similar to other terrestrial biosphere models (McGuire et al., 2001), urban is treated as grassland.

2.3. Model parameterization

In this study, we used Bayesian calibration for model parameterization, which is to determine the optimal value for each parameter in the CH_4 and N_2O modules. A set of major parameters related to CH_4 and N_2O processes were listed with their prior values for simulation (Table 1). Based on these prior parameters and measured site-level fluxes of CH_4 (Table 2) and N_2O (Table 3), we used Monte Carlo method to find the optimal value for each parameter (Robert and Casella, 2005; Ricciuto et al., 2008). The parameters that give the best fit to the observational fluxes were considered as the optimal parameters and used for the regional simulation. Because the site-level climatic data are not always available, we retrieved the site-level data from our regional dataset. We used measurement data of CH_4 and N_2O fluxes from field sites outside North America if the site-specific data of these fluxes for a specific ecosystem type are not available in North America. The sites chosen for model parameterization included 20 sites for CH_4 fluxes (Table 2), and 18 sites for N_2O fluxes (Table 3). Finally, a suite of

parameters (eight for CH_4 module, and four for N_2O module) for each plant functional type were identified for regional model simulation (Tables 4 and 5).

Table 1 Prior estimates of the major parameters for methane and nitrous oxide modules in the Dynamics Land Ecosystem Model (DLEM)

Parameter	Category	Value	Range	Literature
$V_{CH4ProMax}$ (g C m^{-3} day^{-1})	Aerobic	0.0207	0.0033-0.1306	Segers, 1998
	Intermediate	0.4	0.0394-3.9418	Segers, 1998
	Anaerobic	0.75	0.0313-4.9624	Segers, 1998
$V_{CH4OxidairMax}$ (g C m^{-3} day^{-1})		0.10	<0.001-103.7	Sitaula et al., 1995;Segers, 1998;Saari et al., 2004
$V_{CH4OxidtransMax}$ (g C m^{-3} day^{-1})		0.5	0- >51.84	Segers, 1998
$V_{CH4Oxidsoilmax}$ (g C m^{-3} day^{-1})		0.5	0- >51.84	Segers, 1998
$Km_{CH4Prod}$ (g C m^{-3})		10	1.68 – 91.2	Lokshina et al., 2001
$Km_{CH4Oxidair}$ (ppm)		10	5-18	Nedwell and Watson, 1995;Arah and Stephen, 1998;Saari et al., 2004
$Km_{CH4Oxidtrans}$ (g C m^{-3})		5	0.33-19.95	Harrison, 1973;Joergensen, 1985;Linton and Buckee, 1977;Lamb and Garver, 1980;Nagai et al., 1973;O'Neill and Wilktnson, 1977;Ferenci et al., 1975
$Km_{CH4Oxidsoil}$ (g C m^{-3})		10	0.33-19.95	Ferenci et al., 1975;Nagai et al., 1973;Linton and Buckee, 1977;Lamb and Garver, 1980;Joergensen, 1985;Harrison, 1973;O'Neill and Wilktnson, 1977
$V_{max,denit}$ (g N m^{-3} day^{-1})	Natural ecosystems	0.01	0-0.109	Kim et al., 1997;Garcia-Ruiz et al., 1998;Starry et al., 2005
	Cropland	0.05	0-1.0*	
K_{denit} (g N m^{-3})	Natural ecosystems	0.75	0.183-6.5	Garcia-Ruiz et al., 1998;Yu et al., 2006
	Cropland	1.5	1-10£	
$V_{max,nit}$ (g N m^{-3} day^{-1})	Natural ecosystems	0.02	0-2.18	Kim et al., 1997;Sheibley et al., 2003
	Cropland	0.05	0-5*	
K_{nit} (g N m^{-3})	Natural ecosystems	0.75	0.21-1.11	Sheibley et al., 2003
	Cropland	1.5	1-10£	

*assume cropland has two time higher maximum rate for nitrification and denitrification, and higher half-saturation coefficient than natural ecosystems based on *Wang et al's* study (Wang et al., 2009)

£ assume cropland has higher half-saturation coefficient for nitrification and denitrification than natural ecosystems

Table 2 Study sites from which CH_4 and auxiliary data were collected and used in the calibration of the Dynamics Land Ecosystem Model (DLEM)

Site	Vegetation type	Location	Reference
Glacier Lakes Ecosystem Experiment Site *	Subalpine meadow (tundra)	41.33°N, 106.22°W	Mosier et al., 1993
Bonanza Creek Experimental Forest	Boreal broad leaf deciduous forest	64.8°N, 148.0°W	Whalen et al., 1991
Bonanza Creek Experimental Forest	Boreal needle leaf evergreen forest	64.8°N, 148.0°W	Whalen et al., 1991
Saskatchewan boreal forest *	Boreal needle leaf evergreen forest	53.92°N, 104.69°W	Matson, 2008
Konstanz * #	Temperate broad leaf deciduous forest	49,00°N, 8.00°E	Koschorreck and Conrad, 1993
Weiherbach * #	Temperate broad leaf deciduous forest	49.17°N, 8.72°E	Dörr et al., 1993
Changbaishan #	Temperate broad leaf deciduous forest	46.6°N, 128.47°E	Xiao et al., 2004
Changbaishan #	Temperate broad leaf evergreen forest	46.6°N, 128.47°E	Xiao et al., 2004
Gongga Mountain #	Temperate needle leaf evergreen forest	29.0~30.33°N, 101.5~102.25°E	Dong et al., 2003
Pujo * #	Tropical dry forest	1.39°S, 78.00°W	Dörr et al., 1993
Congo river basin * #	Tropical rain forest	1.5°N, 18.0°E	Tathy et al., 1992
Konstanz * #	Temperate mixed forest	49,00°N, 8.00°E	Koschorreck and Conrad, 1993
Sanjiang #	Deciduous shrub	47.69°N, 133.52°E	Song et al., 2009
"Castel Volturno "Nature Reserve * #	Evergreen shrub	40.95°N, 1.55°E	Castaldi and Fierro, 2005
Central Plains Experimental Range	C3 grassland	40.8°N, 104.75°W	Mosier et al., 1996
Central Plains Experimental Range	C4 grassland	40.83°N, 104.7°W	Mosier et al., 2002
Sanjiang #	Herbaceous wetland	47.69°N, 133.52°E	Song et al., 2009
Marcell Experimental Forest *	Woody wetland	47.53°N, 93.47°W	Dise, 1991
High Plains Agricultural Research Laboratory *	Cropland	41.23°N, 103.00°W	Kessavalou et al., 1998
Mojave Desert *	Desert	37°N, 116°W	Strieg et al., 1992

* indicates portions of model-driven forces from regional dataset;
# indicates site outside of the continental North America.

Table 3 Study sites from which N_2O and auxiliary data were collected and used in the calibration of the Dynamics Land Ecosystem Model (DLEM)

Site	Vegetation type	Location	Reference
Glacier Lakes Ecosystem Experiment Site *	Tundra	41.33°N, 106.33°W	Sommerfeld et al., 1993
Saskatchewan boreal forest *	Boreal broad leaf deciduous forest	53.92°N, 104.69°W	Matson, 2008
Saskatchewan boreal forest *	Boreal needle leaf evergreen forest	53.92°N, 104.69°W	Matson, 2008
Changbaishan #	Temperate broad leaf deciduous forest	46.6°N, 128.47°E	Xiao et al., 2004
Changbaishan #	Temperate broad leaf evergreen forest	46.6°N, 128.47°E	Xiao et al., 2004
Gongga Mountain #	Temperate needle leaf evergreen forest	29.0~30.33°N, 101.5~102.25°E	Dong et al., 2003
Rondónia * #	Tropical dry forest	10.5°S, 62.5°W	Garcia-Montiel et al., 2002
Rondónia * #	Tropical rain forest	10.5°S, 62.5°W	Garcia-Montiel et al., 2002
Changbaishan #	Temperate mixed forest	46.6°N, 128.47°E	Xiao et al., 2004
Sanjiang #	Deciduous shrub	47.69°N, 133.52°E	Song et al., 2009
Arid Lands Ecology Reserve * #	Evergreen shrub	46.37°~46.56°N, 119.47°~119.78°W	Mummey et al., 1997
Neimenggu #	C3 grassland	43.03°N, 119.15°E	Huang et al., 2003
Central Plains Experimental Range	C4 grassland	40.83°N, 104.7°W	Mosier et al., 2002
Sanjiang #	Herbaceous wetland	47.69°N, 133.52°E	Song et al., 2009
Saskatchewan boreal forest *	Woody wetland	53.63°N, 106.20°W	Matson, 2008
Arthur Post Farm in Bozeman *	Cropland	45.67°N, 111.15°W	Dusenbury et al., 2008
High Plains Agricultural Research Laboratory *	Cropland	41.23°N, 103.00°W	Kessavalou et al., 1998
Mojave Desert *	Desert	36.82°N,115.92°W	Billings et al., 2002

* indicates portions of model-driven forces from regional dataset;
# indicates site outside of the continental North America.

Table 4 Values of the major parameters for different ecosystem types in methane module after the Bayesian calibration

Major ecosystem type	$V_{CH4ProMax}$ (g C m^{-3} day^{-1})	$V_{CH4OxidairMax}$ (g C m^{-3} day^{-1})	$V_{CH4Oxidtrans}$ (g C m^{-3} day^{-1})	$V_{CH4Oxidsoilmax}$ (g C m^{-3} day^{-1})	$Km_{CH4Prod}$ (g C m^{-3})	$Km_{CH4Oxidair}$ (ppm)	$Km_{CH4Oxidtrans}$ (g C m^{-3})	$Km_{CH4Oxidsoil}$ (g C m^{-3})
Tundra	0.25	0.085	0.1	0.1	10	10	2.5	3
Boreal broad leaf deciduous forest	0.35	0.08	0.1	0.1	10	10	2.5	3
Boreal needle leaf evergreen forest	0.35	0.071	0.1	0.1	10	10	2.5	3
Temperate broad leaf deciduous forest	0.25	0.042	0.2	0.1	15	10	2.5	3
Temperate broad leaf evergreen forest	0.4	0.027	0.1	0.1	15	10	2.5	3
Temperate needle leaf evergreen forest	0.65	0.039	0.1	0.1	15	10	2.5	3
Tropical dry forest	0.5	0.02	0.1	0.1	15	10	2.5	3
Tropical rain forest	0.45	0.015	0.1	0.1	15	10	2.5	3
Temperate mixed forest	0.65	0.048	0.1	0.1	15	10	2.5	3
Deciduous shrub	0.5	0.031	0.25	0.1	15	10	2.5	3
Evergreen shrub	0.25	0.02	0.2	0.1	15	10	2.5	3
C3 grassland	0.5	0.03	0.2	0.1	15	10	2.5	3
C4 grassland	0.6	0.02	0.1	0.1	15	10	2.5	3
Herbaceous wetland	1.45	0.032	5	2.5	5	10	3.5	3.5
Woody wetland	0.55	0.032	5	2.5	5	10	3.5	3.5
Cropland (dry land)	0.4	0.02	0.3	0.35	15	10	10	12
Desert	0.25	0.05	0.25	0.1	15	10	2.5	3

Table 5 Values of the major parameters for different ecosystem types in nitrous oxide module after the Bayesian calibration

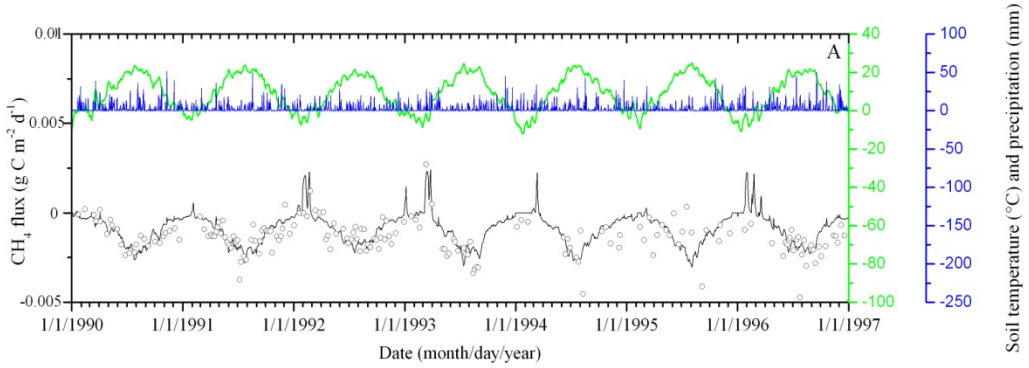
Major ecosystem type	$V_{denimax}$ (g $Nm^{-3}day^{-1}$)	K_{deni} (g Nm^{-3})	V_{nitmax} (g $Nm^{-3}day^{-1}$)	K_{nit} (g Nm^{-3})
Tundra	0.03	0.15	0.008	1
Boreal broad leaf deciduous forest	0.013	0.035	0.0025	1
Boreal needle leaf evergreen forest	0.05	0.05	0.003	1
Temperate broad leaf deciduous forest	0.012	0.15	0.0025	1
Temperate broad leaf evergreen forest	0.007	0.75	0.03	1
Temperate needle leaf evergreen forest	0.012	0.15	0.01	1
Tropical dry forest	0.008	0.25	0.004	1
Tropical rain forest	0.0065	0.15	0.006	1
Temperate mixed forest	0.012	0.15	0.01	1
Deciduous shrub	0.055	0.5	0.005	1
Evergreen shrub	0.16	0.75	0.0025	1
C3 grassland	0.055	0.75	0.005	1
C4 grassland	0.035	0.75	0.0035	1
Herbaceous wetland	0.007	0.5	0.005	1
Woody wetland	0.0013	0.35	0.005	1
Cropland (dry land)	0.052	4.5	0.25	5
Desert	0.01	0.05	0.005	1

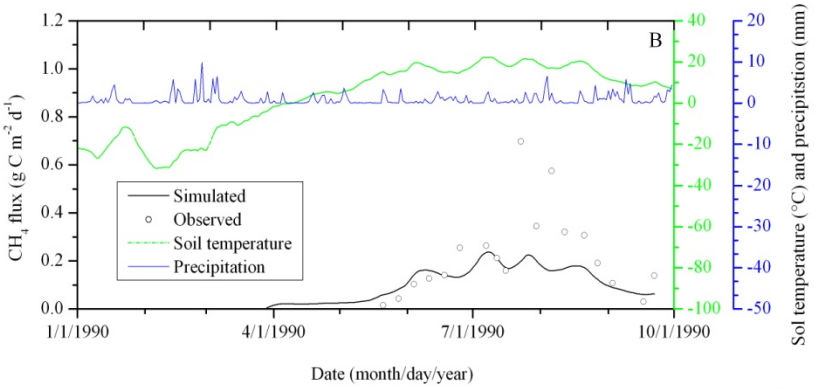
2.4. Model verification

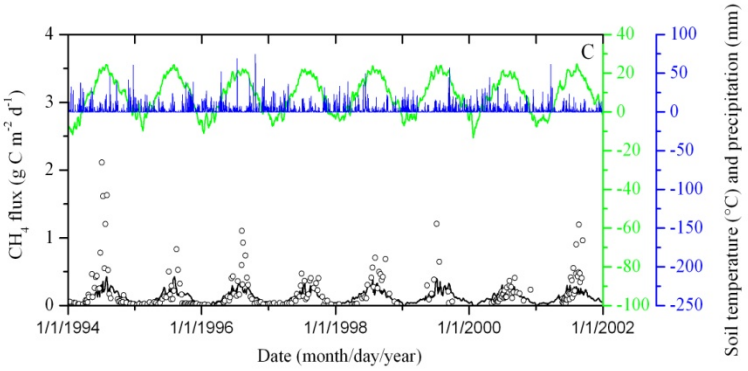
Two forest sites (Durham forest and Hubbard Brook forest) and two wetland sites not used in model parameterization were selected for site-level model verification (Fig. 5). We obtained the observational flux data from various sources including The United States Trace Gas Network (TRAGNET) online dataset (http://www.nrel.colostate.edu/projects/tragnet/), field observations in Hubbard Brook forest by Groffman et al. (2006, 2009) and Sallie fen (Patrick Crill, personal communication, 2008). Four simulations for CH_4 and one for N_2O showed that model results are significantly correlated with observational data even though the DLEM model underestimated some fluxes (Fig. 5A, B, C, D, and E). While the general seasonal patterns of

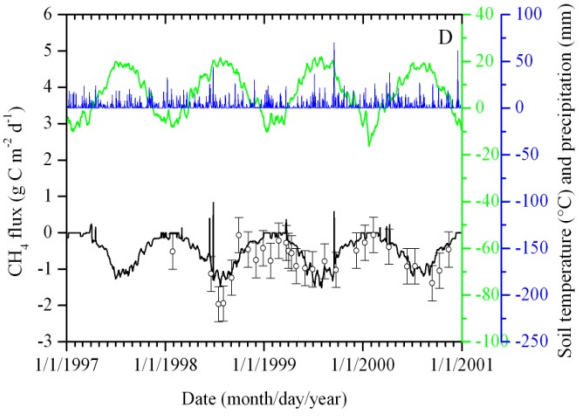
CH_4 flux at these sites were consistent with the observations, the DLEM model did not capture a few CH_4 flux pulses during the peak growing season in the Salle's fen (Fig. 5C), and underestimated CH_4 flux at Alaskan wetland site (Fig. 5 B). For the N_2O flux, the DLEM model well captured the seasonal pattern and annual flux of N_2O in Hubbard Brook forest, but missed several spikes in observational data (Fig. 5E). This phenomenon of peak fluxes in CH_4 and N_2O has been observed in a number of field studies (Chapuis-Lardy et al., 2007; Song et al., 2009), but the underlying mechanisms still remain unknown.

The quantitatively point-to-point comparisons of the modeled and observed data also show that the DLEM captured the seasonal patterns of CH_4 and N_2O fluxes in terrestrial ecosystems at site level. The statistical results could be found in Fig. 5. Comparisons between CH_4 flux with soil temperature and precipitation indicate that the soil temperature is the major factor controlling CH_4 and N_2O fluxes at site-level. The soil temperature is negatively correlated with CH_4 uptake at Durham forest and Hubbard forest sites; while the precipitation events did cause some spikes in CH_4 emission (Fig. 5). For the Alaska wetland and Sallie fen, temperature control on CH_4 emission was obvious, while the precipitation did not show apparent effects on CH_4 emission. For the N_2O emission, temperature effect was shown at seasonal scale, while the precipitation effect appeared at daily scale. This hierarchical control on N_2O emission was consistent with a field study (Brumme et al., 1999).


A
CH4 flux (g C m-2 d-1)
Soil temperature (°C) and precipitation (mm)
Date (month/day/year)
1/1/1990
1/1/1991
1/1/1992
1/1/1993
1/1/1994
1/1/1995
1/1/1996
1/1/1997


B
Simulated
Observed
Soil temperature
Precipitation
CH4 flux (g C m-2 d-1)
Sol temperature (°C) and precipitstion (mm)
Date (month/day/year)
1/1/1990
4/1/1990
7/1/1990
10/1/1990


C
CH4 flux (g C m-2 d-1)
Soil temperature (°C) and precipitation (mm)
Date (month/day/year)
1/1/1994
1/1/1996
1/1/1998
1/1/2000
1/1/2002


D
CH4 flux (g C m-2 d-1)
Soil temperature (°C) and precipitation (mm)
Date (month/day/year)
1/1/1997
1/1/1998
1/1/1999
1/1/2000
1/1/2001

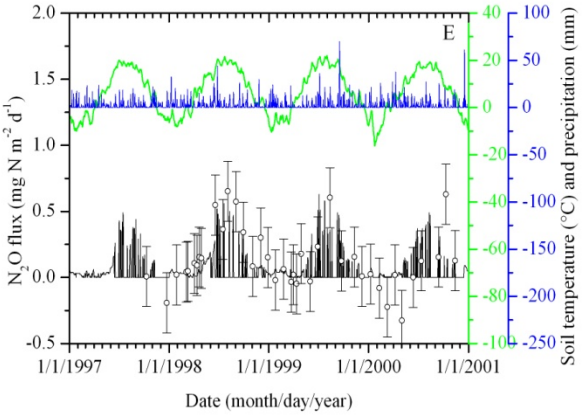

E
N2O flux (mg N m-2 d-1)
Soil temperature (°C) and precipitation (mm)
Date (month/day/year)
1/1/1997
1/1/1998
1/1/1999
1/1/2000
1/1/2001

Fig. 5 Comparison of the DLEM-estimated CH_4 and N_2O fluxes with field observations (A: CH_4 flux in Durham Forest (42˚N, 73˚W) (TRAGNET); B: CH_4 flux in Alaska wetland (64.8˚N, 147.7˚W) (TRAGNET); C: CH_4 flux in Sallie fen (43.21˚N, 71.05˚W) ; D: CH_4 flux in Hubbard Brook Forest (43.95˚N, 71.74˚W) (Groffman et al., 2009; Groffman et al., 2006); E: N_2O flux in Hubbard Brook Forest (43.95˚N, 71.74˚W) (Groffman et al., 2009; Groffman et al., 2006). The error bars in Fig. 4d and 4e represent the standard deviations of four or five replicated observations; the regression models for these five site-level validations are: Modeled = 0.9389 * observed, r = 0.562, P < 0.001 for A; Modeled = 0.5882 * observed, r = 0.628, P < 0.001 for B; Modeled = 0.8795 * observed, r = 0.502, P < 0.001 for C when fluxes higher than 1000 mg C m^{-2} day^{-1} were removed; Modeled = 0.7937 * observed, r = 0.595, P < 0.001 for D; Modeled = 0.7042 * observed, r = 0.633, P < 0.001 for E)

3. Results and Analyses

3.1. Temporal patterns of CH_4 and N_2O fluxes in North America

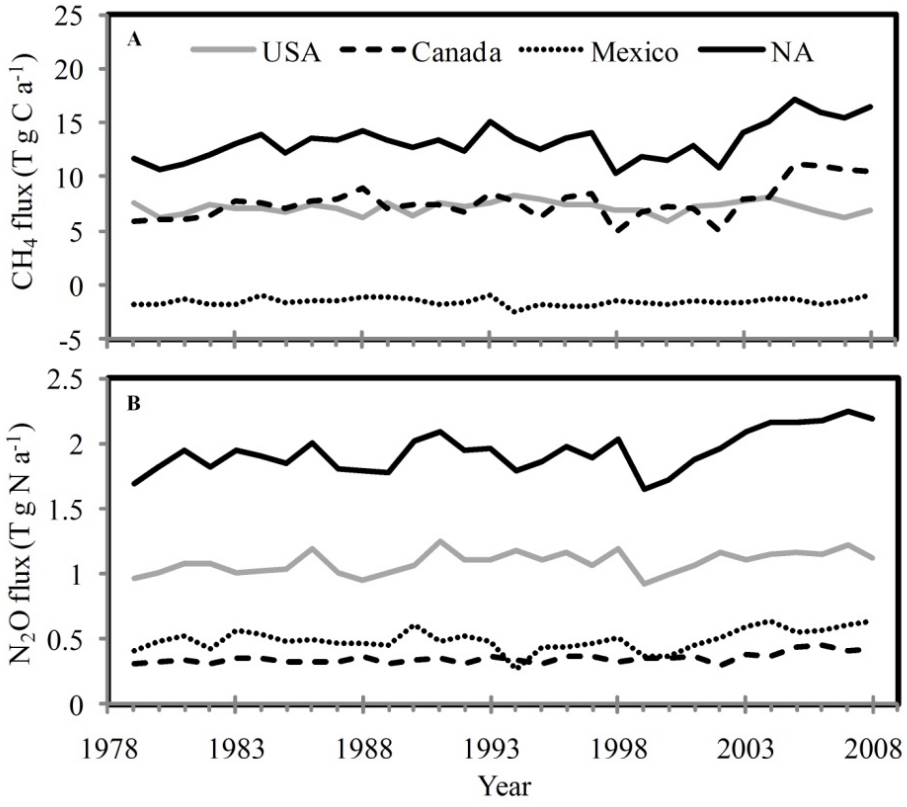


Fig. 6 CH_4 and N_2O fluxes in North America's terrestrial ecosystems by countries during 1979-2008

The annual fluxes of CH_4 and N_2O over the entire North America showed significant fluctuations during 1979-2008. The highest CH_4 emission was 18.42 T g C a^{-1} in 2005, and the lowest was 11.74 T g C a^{-1} in 1998. Before 2001, the annual CH_4 flux was relatively constant with no obvious trend of change; however, since 2002 the CH_4 emission rate increased rapidly, reached the maximum in 2005, and decreased slightly since then (Fig. 6). The overall increasing rate of CH_4 flux was 0.10 T g C a^{-1} over the past 30 years. The highest N_2O emission was 2.25 T g N a^{-1} in 2007, and the lowest was 1.66 T g N a^{-1} in 1999 (Fig. 6). The overall increasing rate of N_2O was 0.01T g C a^{-1} over the past 30 years. The mean annual fluxes over the past 30 years in North America's terrestrial ecosystems were 14.69 ± 1.64 T g C a^{-1} for CH_4, and 1.94 ± 0.16 T g N a^{-1} for N_2O, respectively.

3.2. Spatial distributions of CH_4 and N_2O fluxes in North America

The CH_4 and N_2O fluxes over the entire continent of North America showed significant spatial variations (Fig. 7). The spatial pattern of CH_4 fluxes was mostly dependent on the biome distribution, with a major source located in northwestern part of North America, a region mainly featuring natural wetland. The southwestern part of North America acted as a sink for atmospheric CH_4. A weak sink of CH_4 was also shown in the northeastern part of North America.

All terrestrial ecosystems in North America acted as sources for atmospheric N_2O. The strong sources in southeastern part of North America included the southeastern United States and entire Mexico, where N_2O emission reached as high as 0.8 g N m^{-2} a^{-1}. The weak N_2O sources were observed in other areas, for example, the north part of North America where N_2O was released at a rate of approximately 0.01 g N m^{-2} a^{-1}.

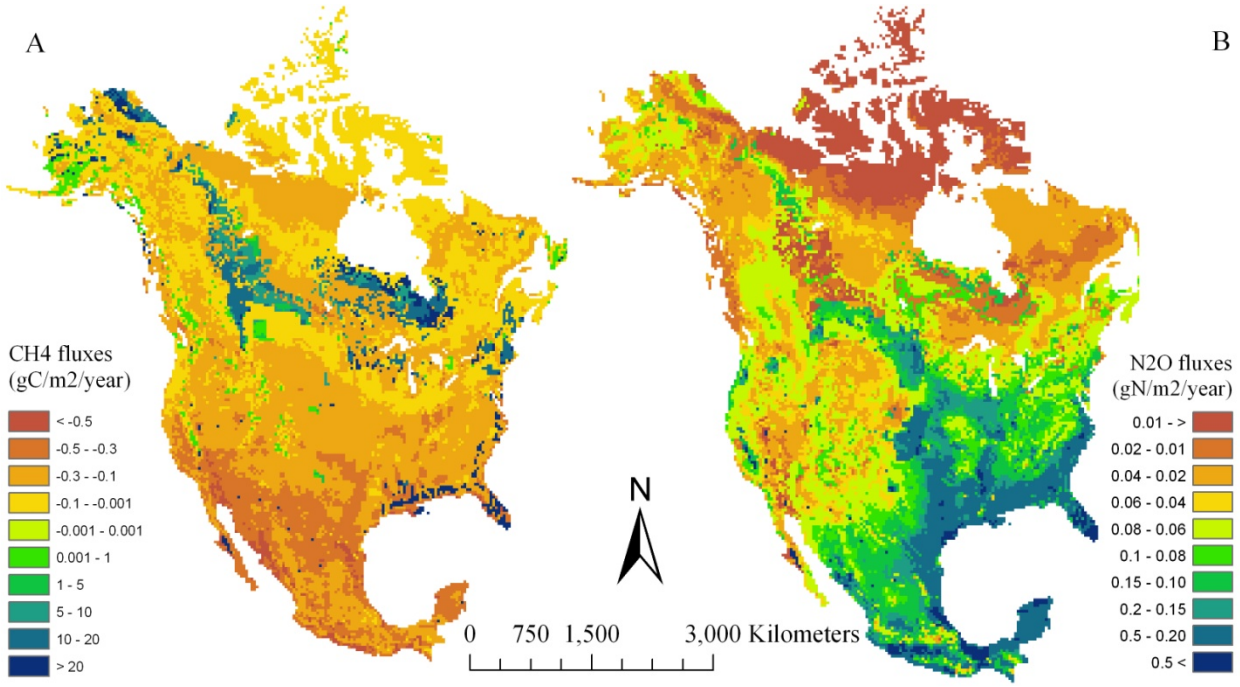


Fig. 7 Spatial distribution of (A) CH_4 and (B) N_2O fluxes in North America's terrestrial ecosystems during 1979-2008

3.3. CH_4 and N_2O fluxes in different countries

The United States, Canada, and Mexico are three diverse countries with different landscapes and anthropogenic activities, resulting in various CH_4 and N_2O flux regimes. At the country level, CH_4 flux was 7.16 ± 0.58 T g C a^{-1} for the United States, 7.68 ± 1.59 T g C a^{-1} for Canada, and -0.15 ± 0.03 T g C a^{-1} for Mexico. The United States and Canada accounted for 48.76% and 52.29%, respectively, and Mexico captured 1.05% of the continental emission of CH_4 (Fig. 6). The country level N_2O flux was 1.09 ± 0.08 T g N a^{-1} for the United States, 0.35 ± 0.04 T g N a^{-1} for Canada, and 0.50 ± 0.08 T g N a^{-1} for Mexico. The United States, Canada, and Mexico accounted for 56.19%, 18.23%, and 25.58%, respectively, of the continental emission of N_2O (Fig. 6).

The rate of changes in CH_4 and N_2O fluxes varied among countries. Based on the regression analysis, we estimated that over the past 30 years, CH_4 emission increased at rates of 5.7 G g C a^{-1} (1 Gg = 10^9 g) in the United States and 91.7 G g C a^{-1} in Canada, while CH_4 consumption increased 0.2 G g C a^{-1} in Mexico. Our results also indicate that N_2O emission increased at rates of 4.2 G g N a^{-1} in the United States, 2.9 G g N a^{-1} in Canada and 2.9 G g N a^{-1} in Mexico, respectively, during the past 30 years.

3.4. CH_4 and N_2O fluxes in different biomes

CH_4 and N_2O fluxes varied remarkably among different ecosystems. Due to the perennial or ephemeral inundated condition, wetlands dominated CH_4 emissions in North America with a source of 17.75 ± 1.63 T g C a^{-1}. All the other ecosystems acted as sinks for atmospheric CH_4 with a total sink of 3.06 ± 0.14 g C a^{-1}. Tundra, forest, grassland, shrub, cropland, and desert and others oxidized atmospheric CH_4 at rates of 0.41 ± 0.03, 1.13 ± 0.07, 0.47 ± 0.03, 0.64 ± 0.02, 0.32 ± 0.03, and 0.10 ± 0.01 T g C a^{-1}, respectively, accounting for -2.79% (negative means uptake), -7.67%, -3.19%, -4.34%, -2.17%, and -0.70% of the continental CH_4 emission (Table 6).

All ecosystems functioned as sources of N_2O. Tundra, forest, grassland, shrub, wetland, cropland, and desert and others emitted 0.07 ± 0.01, 0.63 ± 0.03, 0.22 ± 0.04, 0.25 ± 0.03, 0.19 ± 0.01, 0.56 ± 0.07, and 0.03 ± 0.004 T g N a^{-1}, accounting for 3.68%, 32.21%, 11.24%, 12.72%, 9.78%, 28.82%, and 1.55%, respectively, for the N_2O emission in North America's terrestrial ecosystems (Table 6).

The fluxes of CH_4 and N_2O in each biome over the past 30 years varied significantly. From 1979 to 2008, the CH_4 emission increased at a rate of 103.9 G g C a^{-1} in natural wetland, and the CH_4 oxidation increased at rates of 2.5 G g C a^{-1} in forests, 0.8 G g C a^{-1} in shrub, 0.8 G g C a^{-1} in grassland, and 0.6 G g C a^{-1} in desert and others, respectively. No significant changes

were found for other ecosystem types. For the same time period, the N_2O emission increased at rates of 5.5 G g N a^{-1} in cropland, 1.5 G g N a^{-1} in grassland, 0.8 G g N a^{-1} in tundra, and 0.3 G g N a^{-1} in desert and others. We did not find significant changes for other ecosystem types.

Table 6 Biome contributions to the terrestrial fluxes of CH_4 and N_2O over continental North America (The fluxes are shown as mean plus and minus standard deviation)

		Tundra	Forest	Grassland	Shrub	Wetland	Cropland	Desert and others
CH_4	Flux (T g C a^{-1})	-0.41 ± 0.03	-1.13 ± 0.07	-0.47 ± 0.03	-0.64 ± 0.02	17.75 ± 1.63	-0.32 ± 0.03	-0.10 ± 0.01
	Percentage	-2.79%	-7.67%	-3.19%	-4.34%	120.86%	-2.17%	-0.70%
N_2O	Flux (T g N a^{-1})	0.07 ± 0.01	0.63 ± 0.03	0.22 ± 0.04	0.25 ± 0.03	0.19 ± 0.01	0.56 ± 0.07	0.03 ± 0.004
	Percentage	3.68%	32.21%	11.24%	12.72%	9.78%	28.82%	1.55%

Biome-based estimates may not sum to totals because of the effects of rounding in reporting those estimates

4. Discussion

4.1. Regional comparison to other studies

We estimated annual fluxes of CH_4 and N_2O in terrestrial ecosystems of North America with a spatial resolution of 32km x 32km for the past 30 years. Wetlands predominately account for the continental CH_4 emission. Putting our estimate at the global context (Denman et al., 2007), it accounted for less than 20% of the global CH_4 emissions from natural wetlands at 100 – 231 T g CH_4 a^{-1} (Denman et al., 2007), which is lower than its areal portion of global natural wetland. This may be due to less tropical natural wetlands and rice paddies in this region, which are two strong emitters of CH_4 (Denman et al., 2007; Mitsch and Gosselink, 2007). While our regional estimations of CH_4 and N_2O in North America's terrestrial ecosystems were comparable to previous studies, we found that there were differences at some specific areas or ecosystems. For example, Zhuang et al (2007) used a process-based model (Terrestrial Ecosystem Model) to estimate that the CH_4 emission in Alaska was 2.35 T g C a^{-1} for the period of 1980-1996, which is 12% higher than our estimation of 2.10 T g C a^{-1} for the same time period. However, their estimation of CH_4 emissions in Canada of 5.33 T g C a^{-1} (Zhuang et al., 2004) is 26% lower than our estimation of 7.23 T g C a^{-1} for the 1990s. Combining satellite imageries and a process-based ecosystem model, Potter et al (2006) estimated that CH_4 emission from natural wetland in conterminous United States during 1996-2005 was 4.13 T g C a^{-1}, which is 35% lower than our estimate of 6.34 T g C a^{-1} for the same time period. In addition, a recent synthesis by Bridgham et al (2006) indicated that CH_4 emission in North America's wetlands is 6.75 T g C a^{-1}, which is only 38% of our estimation (17.75 ± 1.63 T g C a^{-1}). Bridgham et al (2006) used site specific estimates of CH_4 fluxes from literature to extrapolate to the wetlands of the entire North America. In *Bridgham et al's* study, they made the simplifying assumption that wetlands, soils

and climate are uniform for each wetland type across the North America for the period of interest. In contract, the DLEM estimates account for spatial variability of wetlands, soils and climate that give rise to place to place differences in CH_4 fluxes over a time period of 30 years. The difference in up-scaling methods used and the time periods examined in the two studies might explain this large discrepancy given the large range of CH_4 flux in different wetland types, soils and climate zones (Bridgham et al., 2006; Barlett and Harriss, 1993; Song et al., 2009). The differences between these estimates were caused largely by the data and models used for their estimations. Using an improved process-based ecosystem model and the most updated and detailed input data, our modeling approach addressed various ecosystem processes and multiple environmental factors that control CH_4 and N_2O fluxes in terrestrial ecosystems.

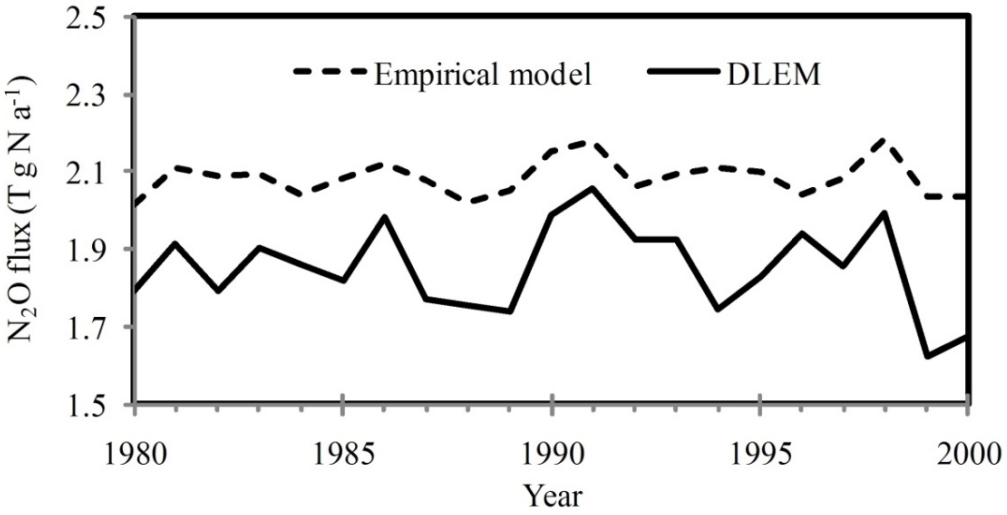


Fig. 8 Comparison of the DLEM-derived N_2O fluxes with the estimations by an empirical model (Xu et al., 2008). The regression model is: the DLEM-derived N_2O = 0.8887 * Empirically-modeled N_2O, R^2 = 0.39; $P < 0.01$.

There are only a few studies of N_2O at large scales (Smith et al., 2004; Liu, 1996; Li et al., 1996; Zhuang et al., 2004; Del Grosso et al., 2005; Xu et al., 2008). The DLEM simulated N_2O emission in North America's terrestrial ecosystems was consistent with a few other

available studies. In the time period of 1980-2000, DLEM-modeled N_2O flux was 1.85 ± 0.11 T g N a^{-1}, comparable to 2.08 ± 0.05 T g N a^{-1} estimated by Xu et al. (2008) (Fig. 8). Inter-annual variations of N_2O fluxes also showed a good agreement between these two studies (R^2 = 0.39; P < 0.01). The spatial correlation of N_2O fluxes between these two studies showed a correlation coefficient of 0.54 (N = 7691). The close agreement between these two studies indicated that DLEM reasonably captured the temporal and spatial patterns of N_2O emission in North America's terrestrial ecosystems. At the same spatial scale and over the same time period, the DLEM-simulated N_2O emission from cropland in United States is higher than *Del Grosso et al*'s estimate for major crops (Del Grosso et al., 2005), but is lower than *Li et al's* estimate for cropland in the continental United States (1996) (Table 7). Putting our estimate at the global context, DLEM-estimated N_2O flux from North America accounted for 20% of the global N_2O source of 9.4 T g N a^{-1} from natural vegetation and agricultural land (Denman et al., 2007). This is proportional to the areal percentage of North America in the global land surface area.

There are also a few inverse estimates on CH_4 and N_2O (Hein et al., 1997; Hirsch et al., 2006; Kort et al., 2008; Chen and Prinn, 2006), and only one study reported the inverse results for natural fluxes from natural wetlands in North America at 9 ± 4.5 T g C a^{-1} (Chen and Prinn, 2006). However, their estimate did not consider the CH_4 flux from lower latitude.

Table 7 Comparison of the DLEM-derived CH_4 and N_2O fluxes with other estimates at the regional scale (DLEM simulations were at a resolution of 32 km × 32 km)

Method	Period	Domain	CH_4 (T g C a^{-1})	N_2O (T g N a^{-1})	Source
Satellite-based empirical method		Wetlands in the continental United States	4.13		Potter et al., 2006
DLEM	1996-2005	Wetlands in the continental United States	6.34±0.43		This study
Process-based model	1990s	Canada	5.33		Zhuang et al., 2004
DLEM	1990s	Canada	7.23±1.11		This study
Process-based model	1980-1996	Alaska	2.35		Zhuang et al., 2007
DLEM	1980-1996	Alaska	2.10±0.27		This study
DNDC at State level	1990	Cropland in the continental United States		0.50-0.74	Li et al., 1996
DLEM	1990	Cropland in the continental United States		0.350	This study
Empirical method at half degree	1980-2000	North America		2.08±0.048	Xu et al., 2008
DLEM	1980-2000	North America		1.85±0.11	This study
DNDC	1970-1999	Agricultural soils in Canada		0.020-0.077 (0.0399) *	Smith et al., 2004
DLEM	1979-1999	Agricultural soils in Canada		0.031-0.055 (0.042) *	This study
DAYCENT at 63 minor regions at county level	1991-2000	Major crop in USA		~0.30#	Del Grosso et al., 2005
DLEM	1991-2000	All crops in USA		0.367±0.048	This study

*Range is shown first and then the mean in bracket
#Data are read from a Figure.

Table 8 Comparison of the DLEM-estimated CH_4 emission rate (g C m^{-2} a^{-1}) with other studies at biome level (Positive values represent CH_4 emission, and negative values represent CH_4 uptake)

Biome type	This study	Bridgham et al., 2006	Barlett and Harriss, 1993	Curry, 2007	Dutaur and Verchot, 2007	Ridgwell et al., 1999
Tundra	-0.101±0.006		-0.055 ~ -0.575	-0.109	-0.112	-0.075
Boreal forest	-0.128±0.010		-0.074 ~ -0.430	-0.125	-0.198	-0.140
Temperate forest	-0.178±0.012		-0.068 ~ -1.15	-0.155	-0.428	-0.181
Tropical dry forest	-0.244±0.013		-0.082 ~ -0.520	-0.199	-0.250	-0.354
Tropical rain forest	-0.221±0.025			-0.202	-0.250	-0.260
Shrub	-0.178±0.006				-0.169	-0.206
Grassland	-0.178±0.010		-0.167		-0.174	-0.238
Desert	-0.185±0.004			-0.129	-0.803	-0.172
Cropland	-0.125±0.014					
Herbaceous wetland	9.985±0.933	Arithmetic: 24.075±5.925 Geometric: 6.075±1.575	26.28 for arctic wetlands; 23.82 for boreal wetlands; 36.96 for temperate bogs and fens; 20.52 for temperate swamps; 19.16 for temperate marshes; 13.14 for temperate floodplains			
Woody wetland	7.871±0.807					

Table 9 Comparison of the DLEM-estimated N_2O emission rate (g N m^{-2} a^{-1}) with other studies at biome level (Positive values represent N_2O emission)

Biome type	This study	Potter et al., 1996	Recalculated from Xu et al., 2008
	Process-based model	Process-based model with remote sensing data	Empirical model
Tundra	0.018 ± 0.002	0.003-0.011	0.002-0.251
Boreal forest	0.047 ± 0.006	0.018	0.016-1.217
Temperate forest	0.107 ± 0.007	0.042-0.064	0.016-1.217
Tropical dry forest	0.110 ± 0.020	0.105	0.175-0.613
Tropical rain forest	0.246 ± 0.039	0.136	0.006-1.060
Shrub	0.061 ± 0.012	0.031	
Grassland	0.094 ± 0.010	0.016	0.004-0.107*
Desert	0.015 ± 0.003	0.004	
Cropland	0.220 ± 0.030	0.081#	0.010-0.725
Herbaceous wetland	0.169 ± 0.014		0.002-0.251
Woody wetland	0.053 ± 0.005		0.002-0.251

* Temperate grassland and tropical savanna and grassland

Without fertilization

4.2. Biome comparison to other studies

The model results showed that wetland ecosystems released CH_4 to the atmosphere while all other ecosystems acted as sinks for CH_4 (Table 8). Herbaceous wetland released CH_4 at a rate of 9.99 ± 0.93 g C m^{-2} a^{-1} and woody wetland at a rate of 7.87 ± 0.81 g C m^{-2} a^{-1}. The strongest sink resided in subtropical/tropical dry forest, and rain forest, followed by grassland, shrub, and desert. The CH_4 emission and consumption in these ecosystems were comparable to other studies (Table 8). For example, the DLEM-estimated CH_4 uptake rate by boreal forest is 0.13 ± 0.01 g C m^{-2} a^{-1}, which is close to *Curry* 's estimate at 0.13 g C m^{-2} a^{-1} (2007), and *Ridgwell et al* 's estimate at 0.14 g C m^{-2} a^{-1} (1999), yet is slightly lower than *Dutaur and Verchot's* estimate at 0.20 g C m^{-2} a^{-1} (2007); the DLEM-estimated CH_4 uptake rate by grassland is 0.18 ± 0.01 g C m^{-2} a^{-1}, which is consistent with *Curry et al* 's estimate at 0.17 g C m^{-2} a^{-1} (2007) and *Dutaur and Vechot* 's estimate at 0.17 g C m^{-2} a^{-1} (2007), yet is slightly lower than *Ridgwell et al* 's estimate as 0.24 g C m^{-2} a^{-1} (1999); the DLEM-estimated CH_4 uptake rate by cropland is 0.12 ± 0.01 g C m^{-2} a^{-1}, which is close to 0.11-0.15 g C m^{-2} a^{-1} (Dobbie et al., 1996; Mosier et al., 1998). The DLEM-estimated CH_4 sink strengths for tundra, temperate forest, shrub, cropland, herbaceous wetland and woody wetland fall in the range of others' estimates (Table 8).

The modeled biome-level fluxes of N_2O are comparable to other studies (Table 9). For almost all biome types, our modeled results are much higher than those estimated by Potter et *al.* (1996), yet in the lower end of the summarized ranges from Xu et al. (2008). For example, the average N_2O flux from tundra is estimated at 0.018 ± 0.002 g N m^{-2} a^{-1} in this study, which is more than 50% higher than *Potter et al*'s estimation at 0.003-0.011 g N m^{-2} a^{-1} (1996); while it is in the lower end of 0.002-0.251 g N m^{-2} a^{-1} summarized in Xu et al (2008). It is same for boreal and temperate forest, shrub, grassland, and tropical rain forest. However, our estimate of N_2O

flux from tropical rain forest is identical to *Potter et al's* estimate (1996) (Table 9). The DLEM-estimated N_2O flux from desert is 0.015 ± 0.003 g N m^{-2} a^{-1}, which is almost three times of *Potter et al's* estimate (1996); however, it is still reasonable comparing with the field observation of 0.04 g N m^{-2} a^{-1} at Sonoran Desert (Guilbault and Matthias, 1998). Our estimated N_2O fluxes from herbaceous and woody wetland are in the middle of summarized range from Xu et al (2008). Given the reported high (Song et al., 2009) or low (Martikainen et al., 1993) N_2O emissions from wetlands, and current state of lacking regional estimation of N_2O from wetlands, further efforts on this issue are highly needed. Emission of N_2O from croplands in the United States in 1990 (0.187 ± 0.139 g N m^{-2} a^{-1}) was also comparable to another estimates of 0.186-0.204 g N m^{-2} a^{-1} by Li et al. (1996).

4.3. Changes in CH_4 and N_2O fluxes among countries

The CH_4 and N_2O fluxes varied substantially among three countries. The Canada had the highest flux and the highest increasing rate in CH_4 emission during 1979-2008, which might be due to its large area of wetland and the high increasing rate of CH_4 production in wetland (Section 3.4.). The highest increasing rate of N_2O emission from the United States was probably caused by large amount of cropland, which was a major source for atmospheric N_2O (Li et al., 1996) (Section 3.3.). Mexico showed relatively high N_2O emission, while acting as a sink for atmospheric CH_4. This decoupling was caused by the landscape composition; the relatively dense cropland made it a major source of N_2O, while small area of wetland emitted little CH_4 to the atmosphere, lower than atmospheric CH_4 consumption by cropland, grassland, and forest (Fig. 4).

4.4. Environmental controls on CH_4 and N_2O fluxes

Inter-annual fluctuation of CH_4 and N_2O fluxes in North America's terrestrial ecosystems was highly correlated with climate factors, especially the mean annual temperature and annual precipitation (Fig. 9). A recent study showed that a drought could reduce N_2O emission from a European spruce forest (Goldberg and Gebauer, 2009); this is consistent with our study, which shows that the droughts in 1994, 1999, and 2002 resulted in relative low N_2O emissions, due to the soil moisture control on denitrification process (see equation 15) (Conrad, 1996). Nitrogen input, including nitrogen deposition and nitrogen fertilizer application, might increase or decrease CH_4 and N_2O fluxes (Steudler et al., 1989; Ding et al., 2004; Liu and Greaver, 2009), while rising atmospheric CO_2 increased CH_4 emission (Hutchin et al., 1995) yet decreased N_2O emissions (Phillips et al., 2001a). Ozone pollution decreased CH_4 emission (Morsky et al., 2008) while increasing or decreasing N_2O emission (Kanerva et al., 2007). The effects of land-cover change are complicated, depending on the direction of the conversion (Willison et al., 1995; Huang et al., 2010). To accurately assess CH_4 and N_2O fluxes in terrestrial ecosystems, it is essential to better understand the underlying mechanisms and attribute the variations in terrestrial ecosystem CH_4 and N_2O fluxes to relative role of various environmental factors.

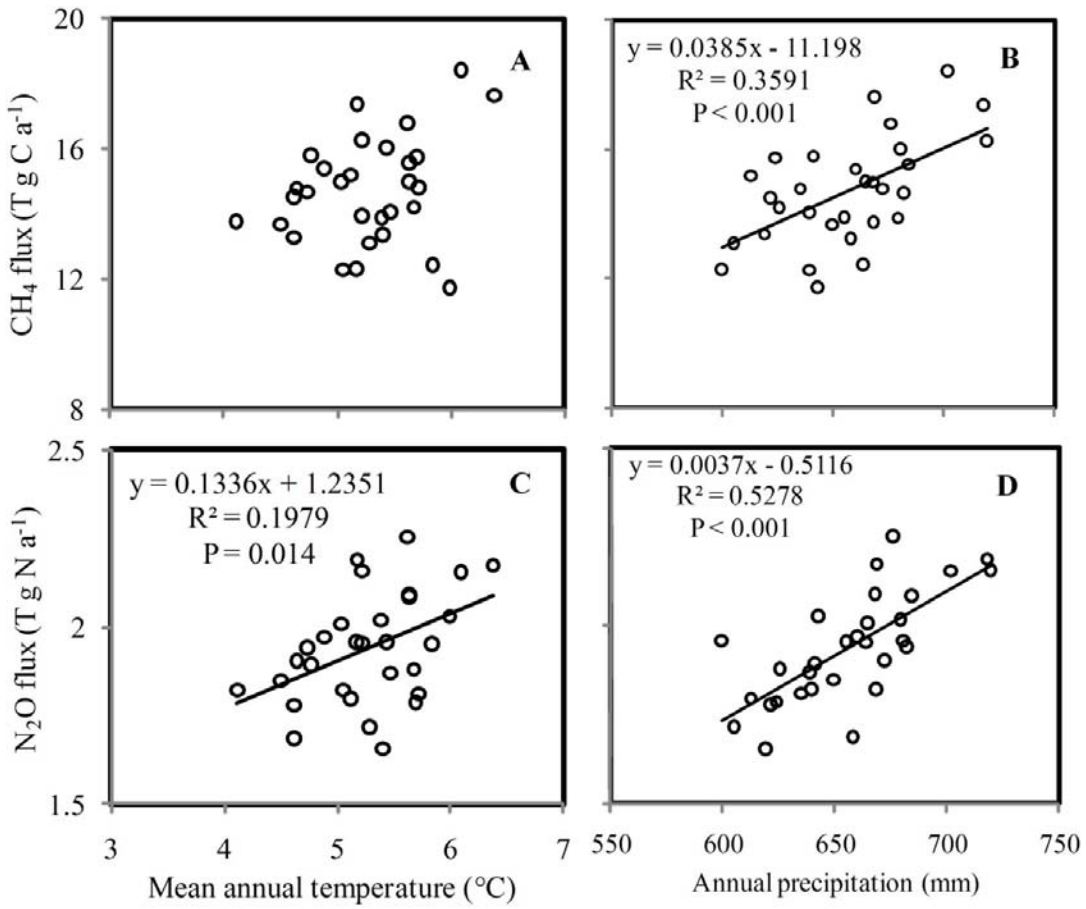


Fig. 9 Correlation between annual CH_4 and N_2O fluxes and mean annual temperature and annual precipitation (A: Correlation between CH_4 flux and mean annual temperature; B: Correlation between CH_4 flux and annual precipitation; C: Correlation between N_2O flux and mean annual temperature; D: Correlation between N_2O flux and annual precipitation)

4.5. Uncertainties in this study and the way forward

We provided regional estimations of CH_4 and N_2O fluxes in North America's terrestrial ecosystems by using an improved process-based biogeochemical model driven by multiple-global change factors. Due to the complexity of the biogeochemical processes related to these two greenhouse gases (Conrad, 1996; Xu et al., 2008), some uncertainties need to be considered when interpreting the results. At the first place, only CH_4 production from DOC was considered in the current model. Other substrates may need to be included, for example, the CH_4 production

from acetate could contribute nearly 20% to CH_4 production (Conrad, 1996; Mer and Roger, 2001). It might be better to include more components in CH_4 production, oxidation, and transport, if these substrates can be quantified. Similarly, improvement of our knowledge and inclusion of more processes in simulating N_2O production and oxidation may be needed. Secondly, current simulation of CH_4 and N_2O fluxes could be underestimated, as the DLEM runs at daily time step and might ignore some possible high pulses in CH_4 and N_2O fluxes at the time step of minute or hour. These high pulses may provide a substantial contribution to the annual fluxes (Brumme et al., 1999). In addition, the availability of soil moisture could turn CH_4 production on and off at the minute or hour time step (Moosavi et al., 1996). Third, the uncertainties in biogeochemical processes and model parameters need to be evaluated. For example, several studies have found the ebullition process might be different from the mechanism applied in the DLEM model (Baird et al., 2004; Kellner et al., 2005; Strack et al., 2005); although these studies pointed out the possible drawback of current representation for this process in process-based model, yet did not provide more reliable method as a replacement. This calls for additional field or experimental investigation before the process can be better addressed in the model. Clearly, parameter uncertainties are highly needed for accurately evaluating the regional CH_4 flux.

Fourth, wetland area and classification could bring uncertainties to regional estimation of CH_4 and N_2O fluxes. The fluxes of CH_4 and N_2O have been reported at an order difference in magnitude among different wetland classes (Barlett and Harriss, 1993; Song et al., 2009), thus the small discrepancy in wetland area and wetland classification could lead to a substantial difference in regional estimation. Fifth, it is important to take into account different mechanisms that control methane production and oxidation in tropical and northern wetlands even though the

mechanisms for the differences between tropical and northern wetlands are still not well documented (Blais et al., 2005). Lastly but not least, N_2O emission from pasture may contribute a great proportion to the continental flux of N_2O (Li et al., 1996). But N_2O emission from managed pasture was not simulated in current model, due to a lack of spatially-explicit information on pasture management.

5. Conclusions

Using the improved Dynamic Land Ecosystem Model (DLEM), we estimated terrestrial ecosystem CH_4 and N_2O fluxes in North America over the past 30 years as a result of multiple global change factors including rising atmospheric CO_2 concentration, ozone pollution, climate change, nitrogen deposition, land-use change, and management. The continental-, country- and biome-level fluxes of CH_4 and N_2O during the past three decades were reported.

This study indicated that approximately 14.69 ± 1.64 T g C a^{-1} (1T g = 10^{12} g) of CH_4, and 1.94 ± 0.16 T g N a^{-1} of N_2O were released from terrestrial ecosystems in North America during 1979-2008. Both the United States and Canada acted as CH_4 sources to the atmosphere, but Mexico mainly oxidized and consumed CH_4 from the atmosphere; and all three countries released N_2O to the atmosphere. Forests and croplands were the two ecosystems that contributed most to continental N_2O emission.

This study provided, to the best of our knowledge, the first continental-level simultaneous quantification and maps at 32 km × 32 km resolution of annual CH_4 and N_2O fluxes in North America's terrestrial ecosystems over the past three decades. While there are some uncertainties related to the estimation of CH_4 and N_2O fluxes due to the simplification of the relevant biogeochemical processes in the model, we believe that this study might provide some useful information for policy making on greenhouse gas mitigation and management. To reduce

uncertainties in regional estimation of CH_4 and N_2O fluxes, it is needed to further improve the representation of additional biogeochemical processes in the DLEM and the spatial datasets of wetland area and pasture management. We also face several key challenges that include attributing the mechanisms responsible for CH_4 and N_2O fluxes and up-scaling from a modeled grid to continental scales.

Chapter 3. Attribution of Spatial and Temporal Variations in Terrestrial Methane Flux over North America[4]

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 ± 9.61 T g CH_4-C (1Tg = 10^{12} g) emission over North America, among which ozone (O_3) pollution led to a reduced CH_4 emission by 2.30 ± 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 ± 12.42, 0.09 ± 0.02, 6.80 ± 0.86, 0.01 ± 0.001, and 3.95 ± 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 ± 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

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negative, on CH_4 emission in Canada; Mexico functioned as a sink for atmospheric CH_4 with a major contribution from elevated atmospheric CO_2. 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 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; Zhuang et al., 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 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.

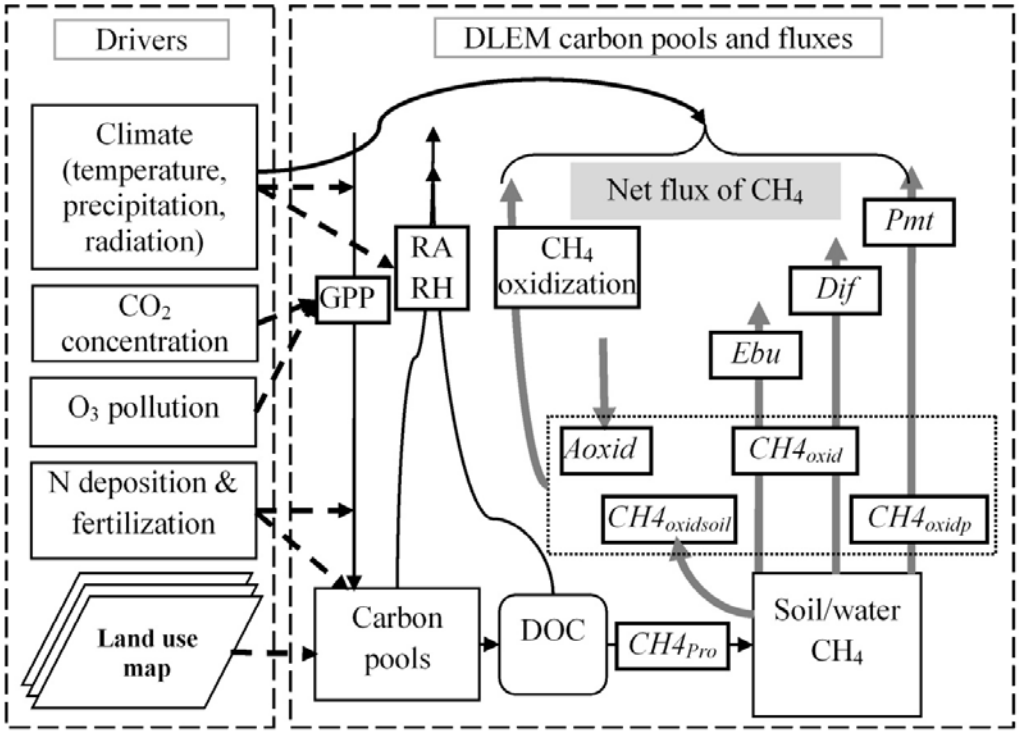
Specifically, the objectives of this study are 1) to examine the factorial contributions to the spatial variation of terrestrial CH_4 flux over North America during 1979-2008; 2) to quantify the factorial contributions to the temporal variations in terrestrial CH_4 flux over North America during 1979-2008; 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, 2010b; 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. 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 et al., 2010), and the CH_4 module has been described in detail in *Tian et al* (Tian et al., 2010a).



Major processes: *Aoxid*: Atmospheric CH_4 oxidation; $CH4_{pro}$: CH_4 production; $CH4_{oxid}$: CH_4 Oxidation during diffusion and ebullition transport; $CH4_{oxidp}$: CH_4 oxidation during plant-mediated transport; $CH4_{oxidsoil}$: 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. 10 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

The methane module in the DLEM model mainly simulates the production, consumption, and transport of CH_4 (Fig. 10). 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., 2010a).

Multiple global change factors yield direct and/or indirect impacts on CH_4 processes as simulated in the DLEM (Fig. 10), which could be expressed as the following equation.

$$\boldsymbol{F_{CH4} = V_{maxprod} f(C_a, w, T_{air}, APAR) f(O_3) f(N) - V_{maxoxid} f(T_{soil}, WFPS)} \quad \text{Equation 24}$$

where F_{CH4} is the CH_4 flux (g C m^{-2} day^{-1}); $V_{maxprod}$ is the maximum rate of CH_4 production (g C m^{-2} day^{-1}); $f(C_a, w, T_{air}, APAR)$ 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 (ppm), w is soil moisture (%); T_{air} is air temperature (°C), $APAR$ is absorbed photosynthetically active radiation (μmol m^{-2} s^{-1}). $V_{maxoxid}$ is the maximum rate of CH_4 oxidation (g C m^{-2} day^{-1}), 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 (°C), $WFPS$ 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 21237 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 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-hour 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.* (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 10). 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 10).

Table 10 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.

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 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. Statistic

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 statistic 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

Table 11 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 ($^{\circ}C \cdot a^{-1}$)	0.04 ± 0.01*
	Minimum temperature ($^{\circ}C \cdot a^{-1}$)	0.03 ± 0.01*
	Average temperature ($^{\circ}C \cdot a^{-1}$)	0.03 ± 0.01*
	Precipitation ($mm \cdot a^{-1}$)	0.65 ± 0.65
	Relative humidity ($\% \cdot a^{-1}$)	-0.01 ± 0.01
	Solar radiation ($W \cdot m^{-2} \cdot a^{-1}$)	0.17 ± 0.03*
Others	O_3 pollution ($ppm\text{-}hr \cdot a^{-1}$)	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*

*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.

Regression analysis was performed to estimate the temporal patterns of major input variables during 1979-2008 (Tables 11, 12, Fig. 11). For the climatic variables, maximum, minimum, and average temperatures, and solar radiation showed significantly increasing rates of 0.04 ± 0.01 $^{\circ}C\ a^{-1}$, 0.03 ± 0.01 $^{\circ}C\ a^{-1}$, 0.03 ± 0.01 $^{\circ}C\ a^{-1}$, and 0.17 ± 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 ± 0.09 $ppm\text{-}hr\ a^{-1}$ 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 fertilizer application, and 1.66 ± 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.

Table 12 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

Spatial variations of input data including potential vegetation distribution, N deposition, N fertilization rate, and O_3 pollution were shown in Fig. 12. The Fig. 12A shows the contemporary spatial distribution of vegetation used in this study; it should be noted that natural wetlands are primarily distributed in Alaska, western Canada, south to the Hudson Bay, eastern coastal area, and Florida in the USA (Fig. 12A). The severely O_3-polluted area over North America locates in western and southeastern parts of North America which could be as high as more than 5000 ppb hr^{-1} (monthly accumulated hourly O_3 dose over a threshold of 40ppb in ppb-hr), while the other areas, especially northern end of continental North America, feature low O_3 pollution (Fig. 12B). 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. 12C). 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. 12D).

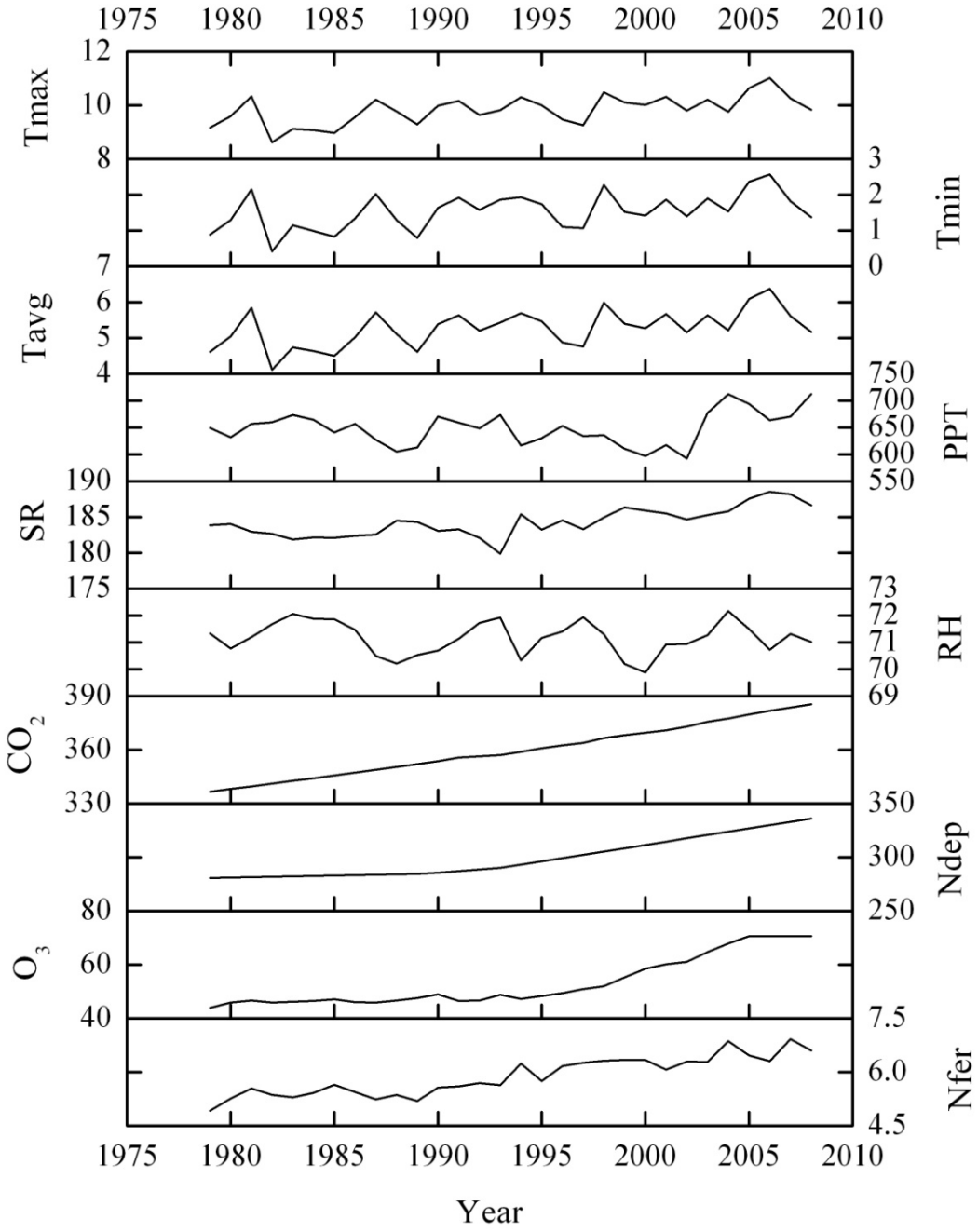


Fig. 11 Temporal dynamics of the variables from 1979 to 2008 (*Tmax*: maximum temperature (˚C); *Tmin*: minimum temperature (˚C); *Tavg*: average temperature (˚C); *PPT*: precipitation (mm); *SR*: solar radiation ($W \cdot m^{-2}$); *RH*: relative humidity (%); CO_2: atmospheric CO_2 concentration (ppm); *Ndep*: N deposition ($mg\ N \cdot m^{-2}$); *O_3*: daily O_3 pollution (ppb-hr); *Nfer*: N fertilization ($g\ N \cdot m^{-2}$))

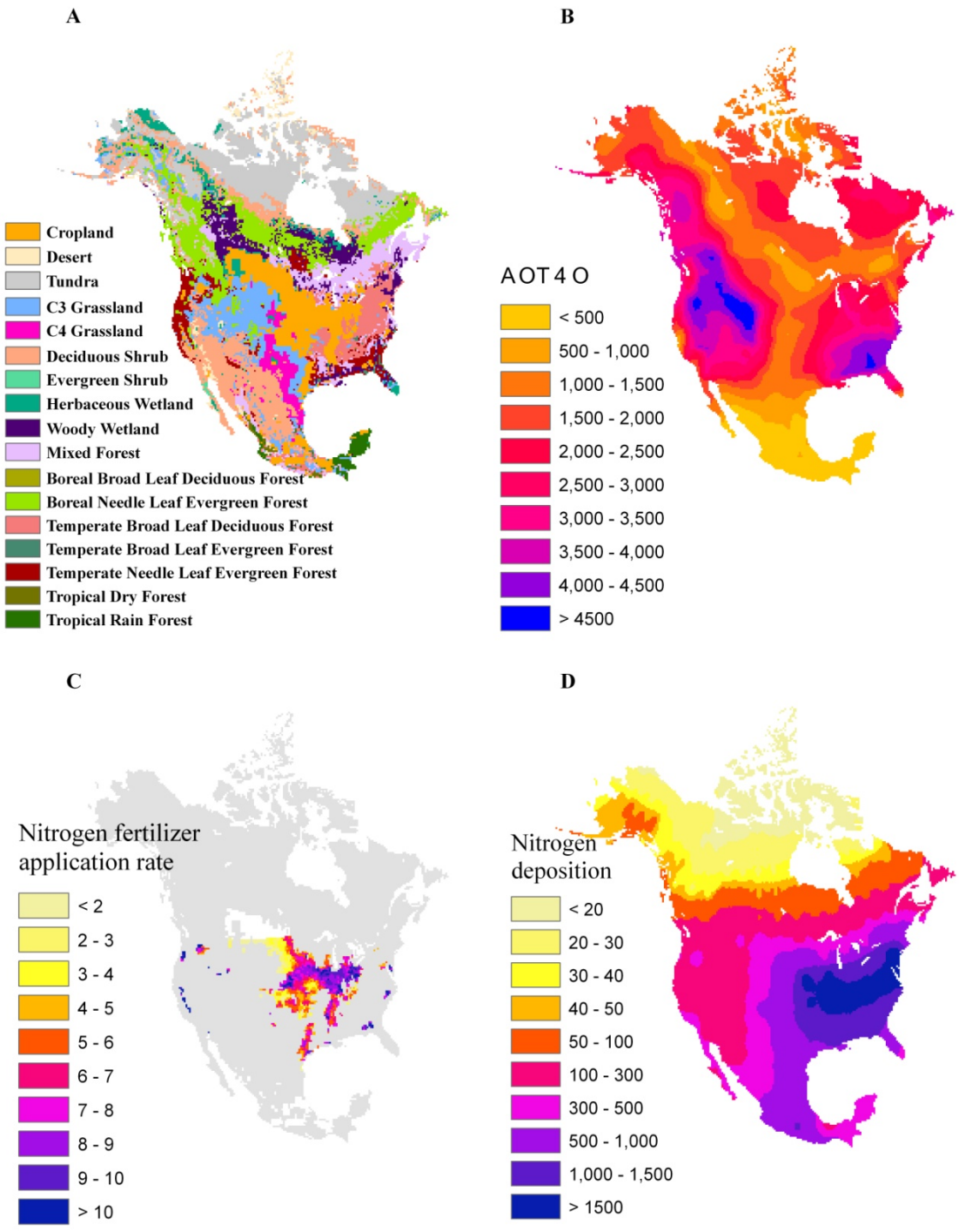


Fig. 12 (A) Contemporary vegetation map, and spatial distribution of 30-year averages of (B) monthly O_3 pollution (AOT40 at ppb/hr), (C) N fertilization application (g N m^{-2} a^{-1}), and (D) N deposition rate (mg N m^{-2} a^{-1})

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. 13); 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.

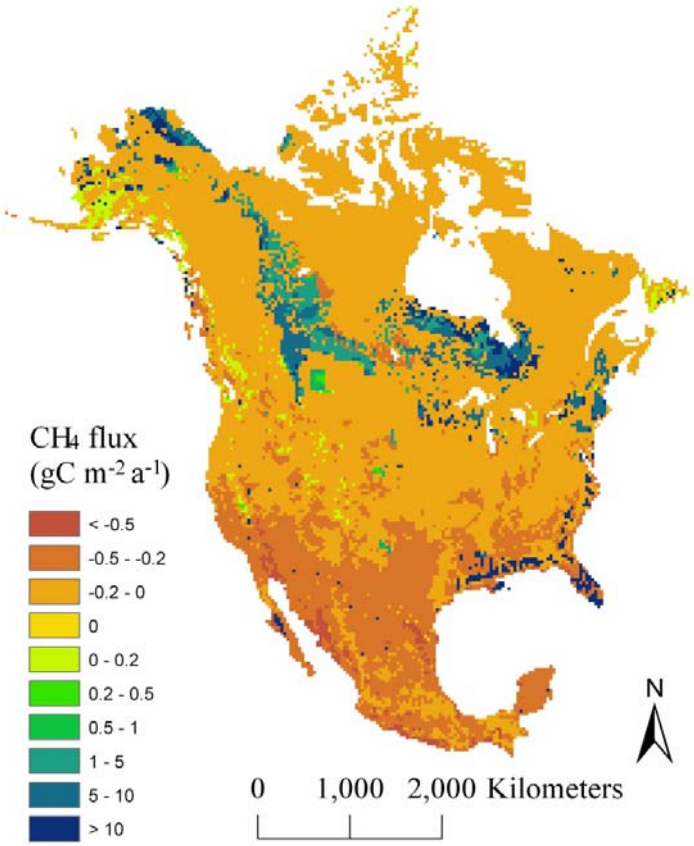


Fig. 13 Spatial variations of terrestrial CH_4 fluxes caused by global change factors over North America from 1979 to 2008

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. 14A); N deposition enhanced CH_4 emission across large area of North America, primarily in eastern parts of Canada, and southeastern US (Fig. 14B); 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. 14C); O_3 pollution exerted no significant effects on CH_4 flux across majority of North America, while decreased CH_4 emission in southeastern parts of the continental North America and enhanced CH_4 emission in small magnitude over portions of Canada (Fig. 14D); N fertilizer application and land conversion slightly enhanced CH_4 emission in portions of agricultural land throughout North America (Fig. 14E & F); interactive effects between/among global change factors enhanced CH_4 emission in large area of North America, especially the Southwest (Fig. 14H); 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. 14G).

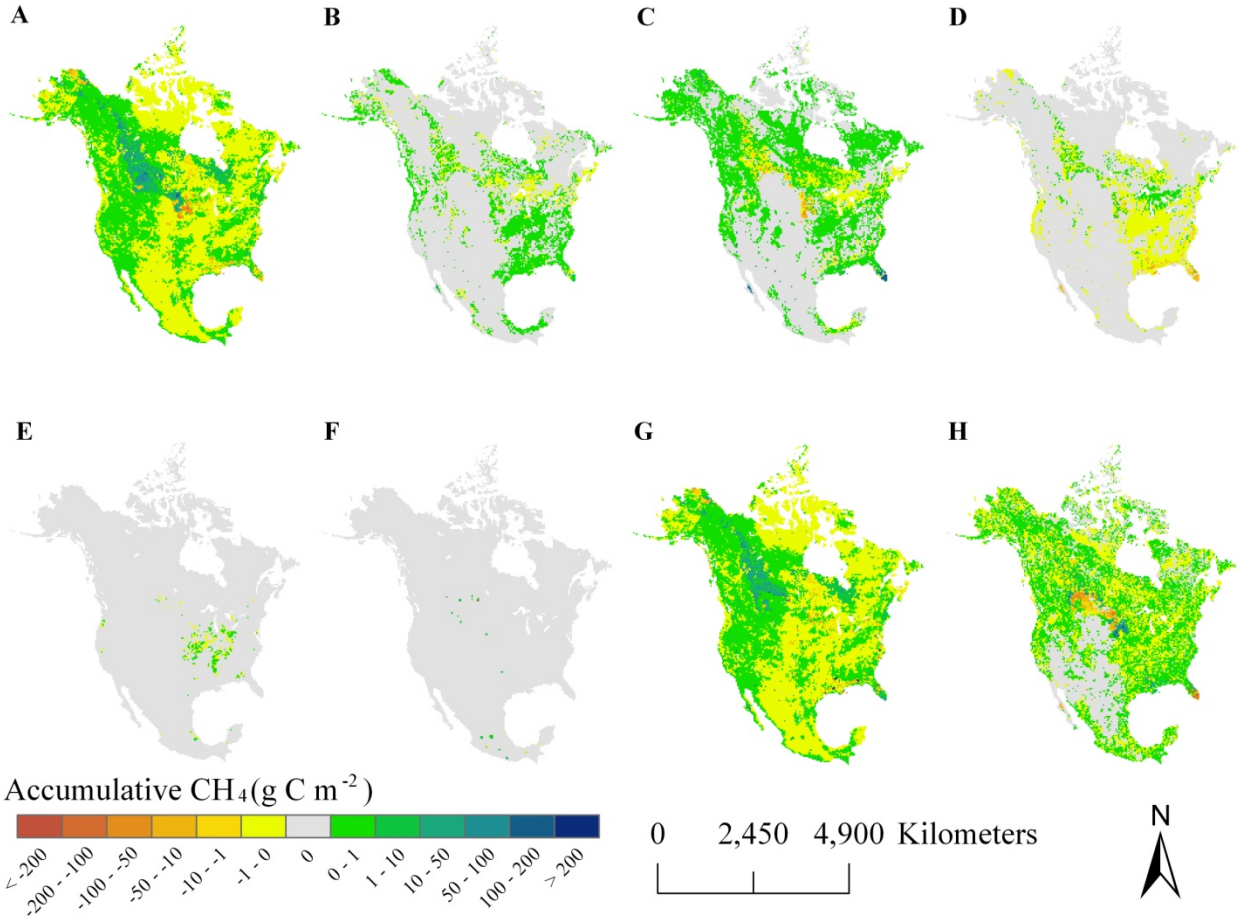


Fig. 14 Factorial contributions to the spatial variations in accumulated CH_4 flux over North America from 1979 to 2008 (A: climatic variability; B: N deposition; C: CO_2; D: O_3 pollution; E: N fertilizer application; F: land conversion; G: all combined; H: interaction)

3.4. 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 during 1979-2008 (Fig. 15). 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 ± 1.64 T g CH_4-C a^{-1}; and the overall increasing rate of CH_4 flux was 0.10 ± 0.03 T g CH_4-C a^{-1} over study period ($P = 0.003$) (Fig. 15A).

Various global change factors yielded significantly different effects on the long-term trends of continental CH_4 flux during 1979-2008 (Fig. 15). Climate variability generated a substantially inter-annual variation in CH_4 flux, with an increasing rate of 0.15 ± 0.04 T g CH_4-C a^{-1} ($P = 0.002$) (Fig. 15B). The continuously rising atmospheric CO_2 concentration kept accelerating CH_4 emission at an overall increasing rate of 0.02 ± 0.004 T g CH_4-C a^{-1} ($P < 0.001$) (Fig. 15D), while O_3 pollution decreased CH_4 emission at a rate of 0.01 ± 0.001 T g CH_4-C a^{-1} ($P < 0.001$) (Fig. 15E). N deposition generated an increasing rate of 0.71 ± 0.05 G g CH_4-C a^{-1} (1Gg = 10^9 g) to continental-level CH_4 emission ($P < 0.001$) (Fig. 15C), while N fertilizer application alone did not exert any significant effects on CH_4 flux at the continental scale (Fig. 15F). 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 ± 0.001 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. 15G). 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 ($P < 0.001$).

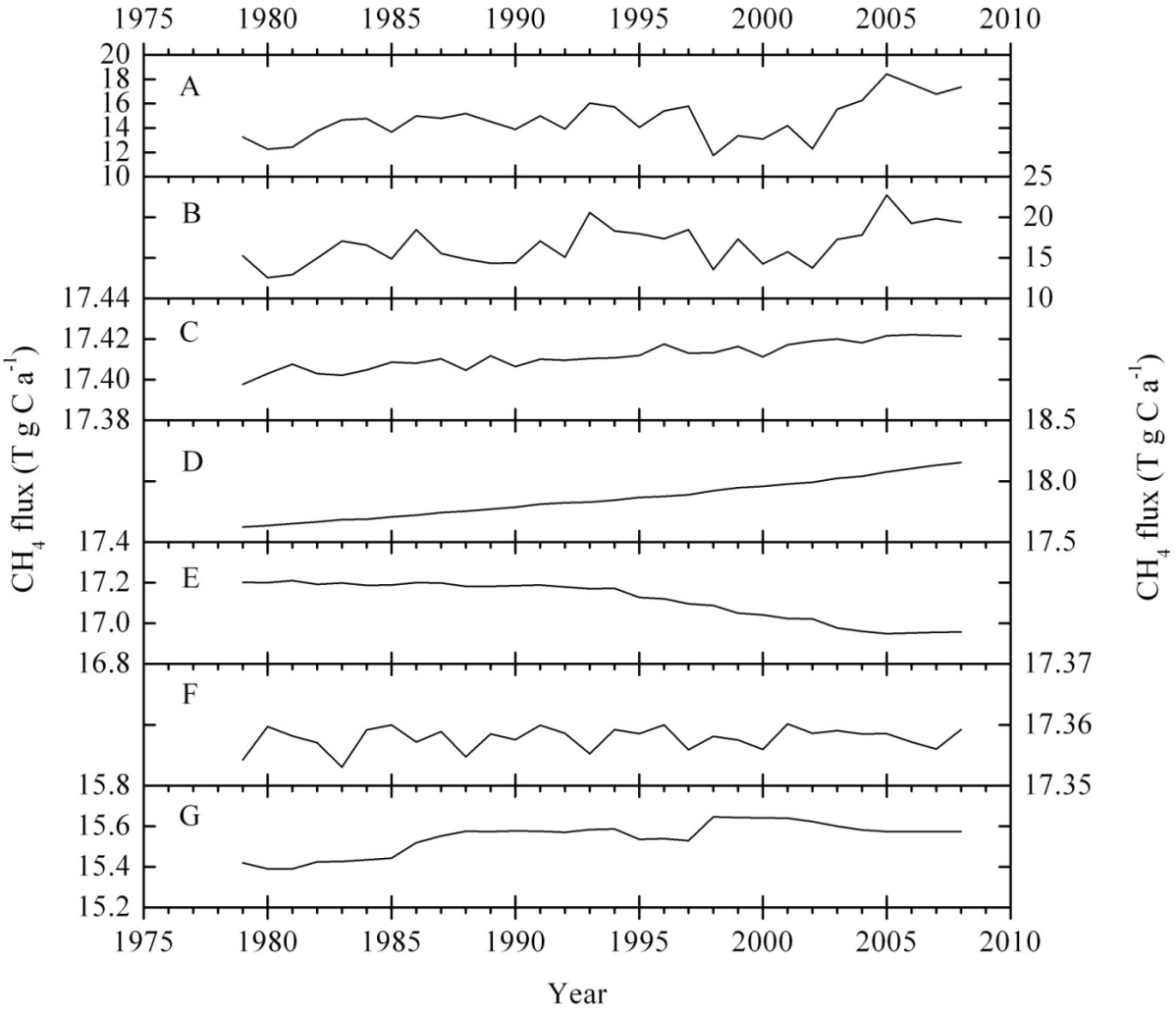


Fig. 15 Temporal variations of terrestrial CH_4 flux caused by global change factors over North America from 1979 to 2008 (A: all combined simulation; B: climate only simulation; C: N deposition only simulation; D: CO_2 only simulation; E: O_3 only simulation; F: N fertilization simulation; G: land conversion only simulation)

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 ± 8.97 T g CH_4-C, of which 417.24 ± 6.83 T g CH_4-C was contributed from baseline flux and 23.51 ± 9.61 T g CH_4-C was caused by global change factors (Table 13). O_3 pollution and the interactive effects between/among multiple factors decreased CH_4 emission by 2.30 ± 0.49 T g CH_4-C and 4.84 ± 7.74 T g CH_4-C, respectively, while all the other single factors increased CH_4 emission from North America's terrestrial ecosystems (Fig. 16).

The 30-year accumulated CH_4 emission was 214.89 ± 3.19 T g CH_4-C for USA and 230.47 ± 8.72 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 ± 0.19 T g CH_4-C over the past 30 years (Table 13). For USA, climate variability and O_3 pollution accumulatively decreased CH_4 emission by 3.49 ± 9.33 T g CH_4-C and 2.06 ± 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 13). For Canada, it is estimated that climate variability accumulatively enhanced CH_4 emission by 23.32 ± 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 13). 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 ± 0.20 T g CH_4-C in Mexico during 1979-2008 (Table 13).

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 14). 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. 17).

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 13).

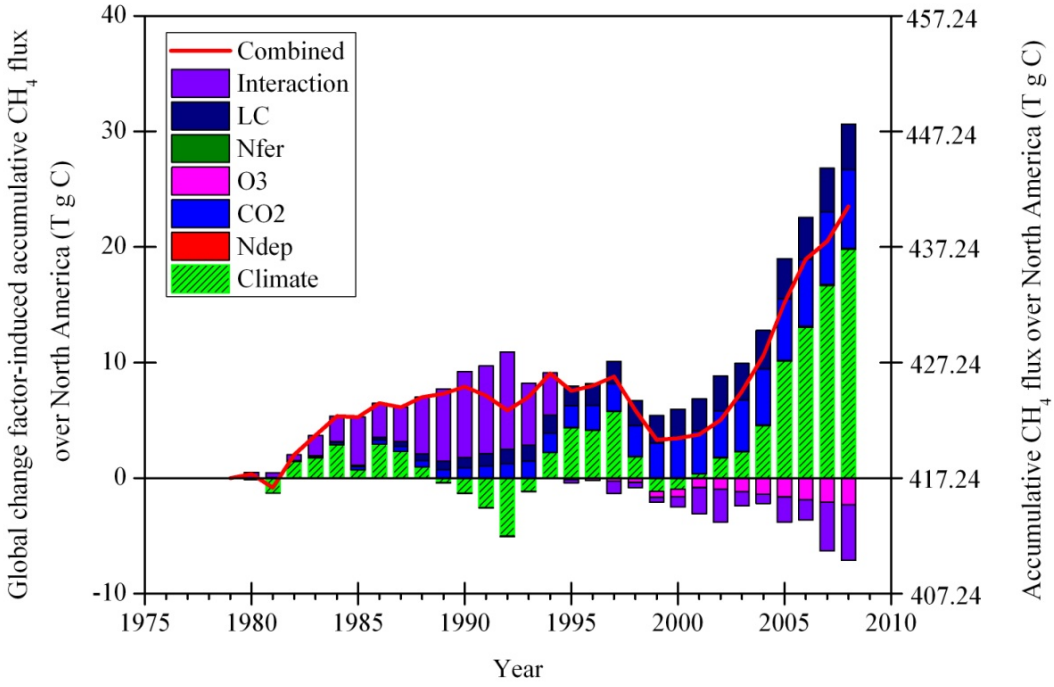


Fig. 16 Factorial contributions to accumulated CH_4 flux over North America during 1979-2008 (The right Y-axis shows the accumulated CH_4 flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from 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)

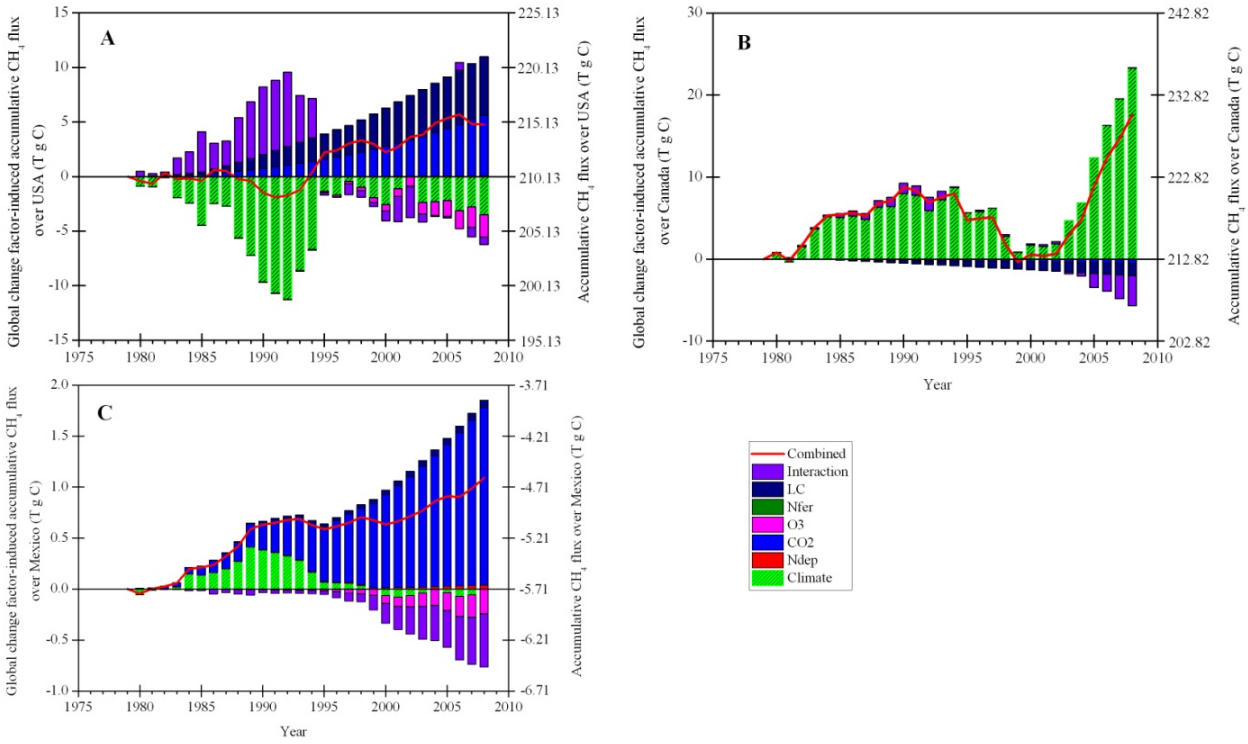


Fig. 17 Factorial contributions to accumulated CH_4 flux at country-level during 1979-2008 (A: USA; B: Canada; C: Mexico) (The right Y-axis shows the accumulated CH_4 flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* 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)

Table 13 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

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

Table 14 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

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

3.6. Factorial contributions to the inter-annual variations in CH_4 flux during 1979-2008at 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. 15). After removing the baseline emission of CH_4, we identified the major factors for the year-by-year variation in CH_4 flux (Fig. 18). 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. 18). 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. 19A). Climate variability outweighed other factors in controlling the increases in terrestrial CH_4 emission over Canada (Fig. 19B). 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. 19C). Further analysis showed that all climate variables made significant contributions, with the largest contribution from precipitation in the USA, and Canada, and temperature in Mexico.

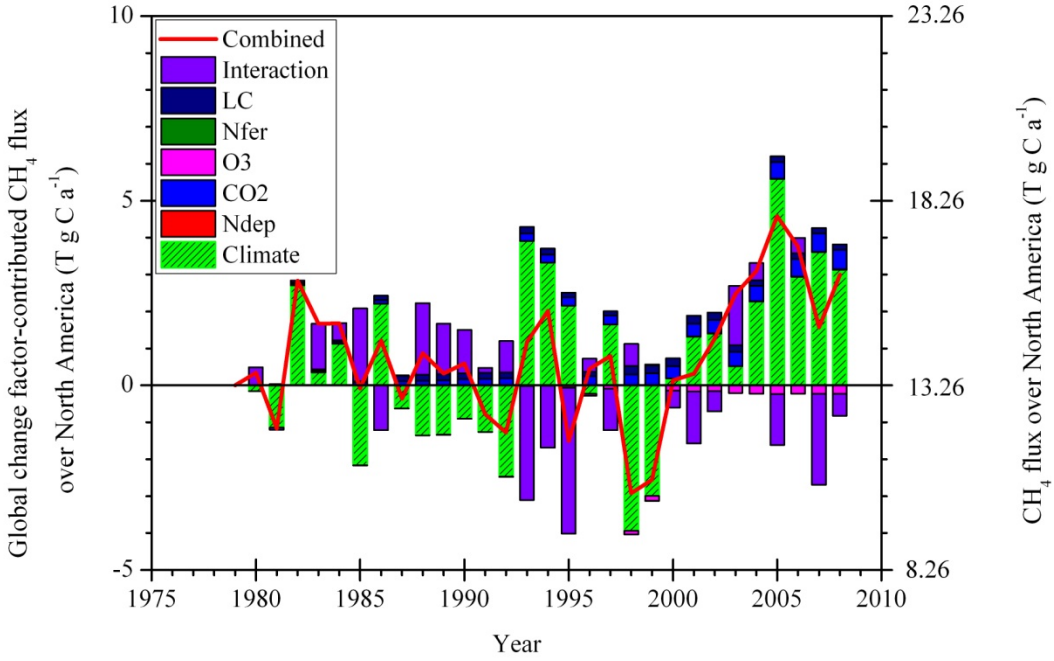


Fig. 18 Factorial contribution to the inter-annual variations in CH_4 flux over North America (The right Y-axis shows the accumulated CH_4 flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from 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)

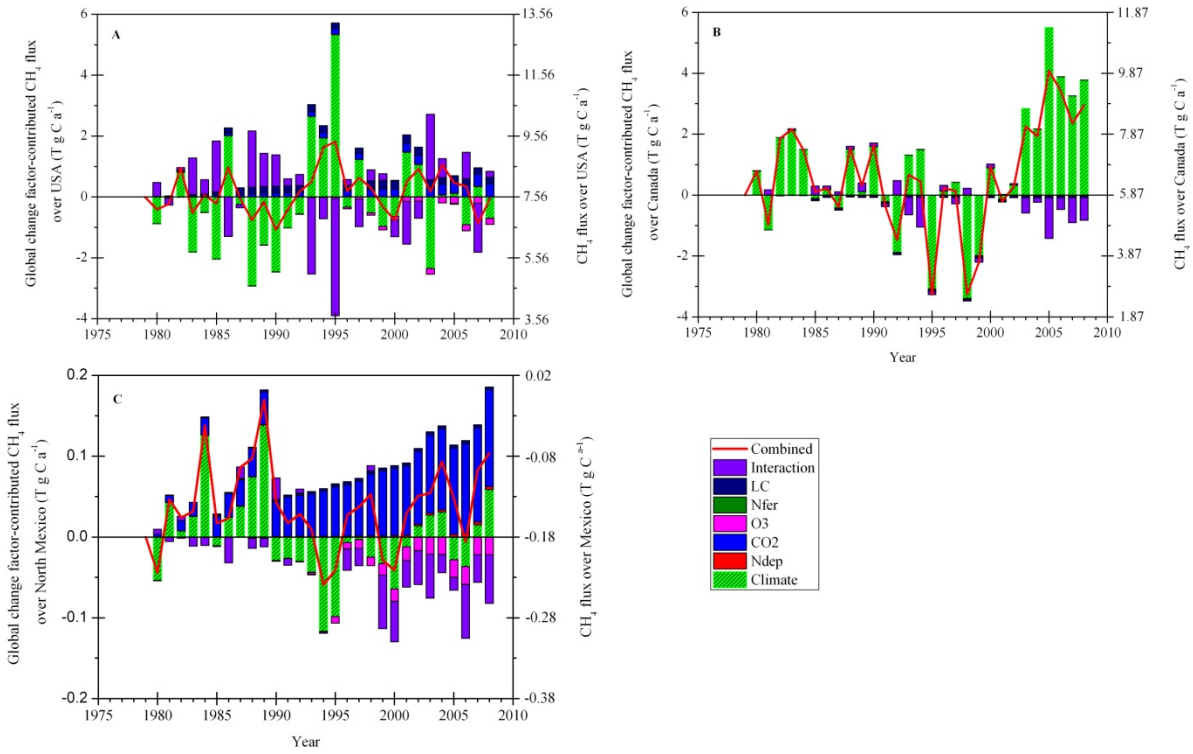


Fig. 19 Factorial contribution to the inter-annual variations in CH_4 flux by country (A: USA; B: Canada; C: Mexico) (The right Y-axis shows the accumulated CH_4 flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* 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)

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 (Fig 11, 12). 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. 12A) (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 15). 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 0~146%, 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% 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 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 15).

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 ± 1.09 mg C m^{-2} a^{-1} per g N^{-1} m^{-2} a^{-1} for dry cropland, comparing to 12 ± 6 mg C m^{-2} a^{-1} per 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 ± 0.02 mg C m^{-2} a^{-1} per g N^{-1} m^{-2} a^{-1} for 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 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 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 ± 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 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* 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 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).

Table 15 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 200ppm increase from 355ppm to 550ppm	+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	360ppm 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 reaver, 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	0 (mg C g N^{-1} m^{-2} a^{-1}) in CH_4 uptake	Liu and Greaver, 2009

			uptake		
	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	10-20ppb 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 ($Y = 0.047 * X – 568.82$, $R^2 = 0.959$, $N = 30$)
£ 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 & 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.

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., 2006; Reich et al., 2001); 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. 16, 18). 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 global circulation models (Forster et al., 2007).

The inter-annual variability in the continental CH_4 flux was dominated by climatic variability (Table 13); this would be supported by the significantly positive correlation between climate-induced and overall CH_4 fluxes (Fig. 15), and the detailed analysis of factorial contribution to terrestrial CH_4 flux over the 30 years (Fig. 16 & 17). 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 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. 16 & 18); 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 14). 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 14).

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 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., 2006;Walter et al., 2007), which may

contribute to the terrestrial CH_4 budget, especially from inland small lake or river (Walter et al., 2006;Walter et al., 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.

Chapter 4. Multiple-factor Control on Terrestrial Nitrous Oxide Flux over North America

Abstract

Nitrous oxide (N_2O) is a potent greenhouse gas which also contributes to the depletion of stratospheric ozone (O_3). However, the magnitude and underlying mechanisms for the spatiotemporal variations in the terrestrial sources of N_2O are still far from certain. Using a process-based ecosystem model (Dynamic Land Ecosystem Model), driven by multiple global change factors including climate variability, nitrogen (N) deposition, rising atmospheric CO_2, O_3 pollution, N fertilization, and land use conversion, the spatial and temporal variations in terrestrial N_2O flux over North America were examined and attributed to various driving factors. From 1979 to 2008, the North America accumulatively emitted 58.17 ± 0.85 T g N_2O-N (1T g = 10^{12} g), of which global change factors contributed 2.81 ± 0.98 T g N_2O-N, and baseline emission contributed 55.35 ± 0.56 T g N_2O-N. The elevated CO_2 led to decreases in terrestrial N_2O emission at 0.51 ± 0.07 T g N_2O-N. Climate variability, N deposition, O_3 pollution, N fertilization, and land use conversion increased N_2O emission by 0.56 ± 0.68, 0.50 ± 0.07, 0.10 ± 0.02, 0.92 ± 0.09, and 0.16 ± 0.01 T g N_2O-N, respectively. The interactive effect among multiple factors enhanced N_2O emission by 1.10 ± 0.37 T g N_2O-N over the 30 years. At country level, climate variability and elevated atmospheric CO_2 decreased, while all other single factors and multiple-factor interaction enhanced N_2O emission in the United States of America (USA) over the past 30 years; during the same time period, elevated atmospheric CO_2 and multiple factor interaction decreased, while other factors enhanced N_2O emission from Canada; and

elevated atmospheric CO_2 and land conversion decreased, while other factors enhanced N_2O emission from Mexico. Southeastern part of the continental North America including central Canada, southeastern USA, and entire Mexico acted as a strong source for atmospheric N_2O while other parts functioned as a weak source of N_2O. The effects of climate variability and multiple-factor interaction dominated the inter-annual variations in terrestrial N_2O emission at both continental and country levels, indicating that the projected change in global climate system during this century may substantially alter the regime of N_2O emission from terrestrial ecosystems. It also implies that the interactive effect among global change factors may yield significant effects on N_2O flux and need more examination with field experiments.

1. Introduction

Nitrous oxide (N_2O) plays an important role in both contributing to greenhouse effect (Denman et al., 2007; Rodhe, 1990) and depleting stratospheric ozone (O_3) (Denman et al., 2007; Cicerone, 1987). The atmospheric N_2O concentration has increased from 270 ppb (one part per billion) in 1750 to 319 ppb in 2005 (Forster et al., 2007); terrestrial ecosystems under the impacts of anthropogenic activities have been recognized as one of major sources for this increase (Keller et al., 1986; Bouwman et al., 1993; del Grosso et al., 2006; Li et al., 1996; Liu, 1996; Repo et al., 2009; Williams et al., 1992; Forster et al., 2007; Denman et al., 2007). Estimating the terrestrial courses of N_2O and examining their factorial contributions will be crucial for advancing our understanding of the dynamics of atmospheric N_2O concentration, and further provide helpful information for policy-making to curb the continuous increase in atmospheric N_2O concentration (Denman et al., 2007; Tian et al., 2009b).

N_2O flux has been recognized as a result of a suite of microbial processes which were influenced by a variety of environmental factors (Conrad, 1996; Williams et al., 1992). Globally changing environments will alter these factors and substrates, and further change the N_2O flux (Bouwman et al., 1993; Conrad, 1996; Goldberg and Gebauer, 2009; Kanerva et al., 2007; Kettunen et al., 2005; Williams et al., 1992; Ambus and Robertson, 1999). For example, nitrogen (N) input may increase N_2O production by increasing substance availability (Kettunen et al., 2005; Mcswiney and Robertson, 2005); elevated atmospheric CO_2 may reduce N availability in soil owing to progressive N accumulation in plant biomass (Luo et al., 2004; McGuire et al., 1995), which cut down the N_2O emission (Phillips et al., 2001); alternatively, elevated atmospheric CO_2 might increase photosynthetic products and stimulate microbial process, and thus increase N_2O emission (Kettunen et al., 2005; Ineson et al., 1998); if these two effects are counterbalanced, it may appear as neutral response of N_2O flux to elevated atmospheric CO_2 (Kanerva et al., 2007; Ambus and Robertson, 1999); O_3 pollution may alter microbial community (Kanerva et al., 2008) and cause increase or decrease in N_2O emission, depending on time and location (Kanerva et al., 2007); the effects of climate variability and land conversion on the N_2O emission is more complicated and really dependent on the specific site condition (Jiang et al., 2009; Goldberg and Gebauer, 2009; Zhang et al., 2007b).

In the past decades, considerable effort has been put on the accurate estimation of terrestrial N_2O flux (Potter et al., 1996; Xu et al., 2008; Liu, 1996; Denman et al., 2007; Matson and Vitousek, 1990; Bouwman et al., 1993). A number of estimates for N_2O flux have been achieved by extrapolating average fluxes from chamber-based measurements to the areal extent of vegetation or soil classes from which the measurements were taken (Keller et al., 1986; Huang et al., 2003; Matson et al., 1989), or by using a simple empirical model (Xu et al., 2008). In these

approaches, uncertainties were introduced because the spatial heterogeneity within the classes, as well as the seasonal and interannual variability in climatic and biotic controls on emission rates were neglected, at least partially (Potter et al., 1996; Matson et al., 1989). Meanwhile, the empirical methods in estimating regional N_2O flux could not be used to attribute the spatiotemporal variations in terrestrial N_2O flux to underlying mechanisms. So, a large-scale estimation of terrestrial N_2O flux with consideration of spatial heterogeneity of soil, vegetation, and climate variations, and also could be used for factorial attribution, is highly needed.

Process-based modeling approach has become more and more popular in conducting regional estimation of N_2O flux (Del Grosso et al., 2006; Li et al., 2001; Potter et al., 1996). Although the process-based models have the potential to explore the potential contribution from each driving forces to the spatiotemporal variations in terrestrial N_2O as they have been conducted for CO_2 flux (McGuire et al., 2001; Tian et al., 2003; Mu et al., 2008), none has been utilized to attribute the spatial and temporal variations in terrestrial N_2O flux to its driving factors.

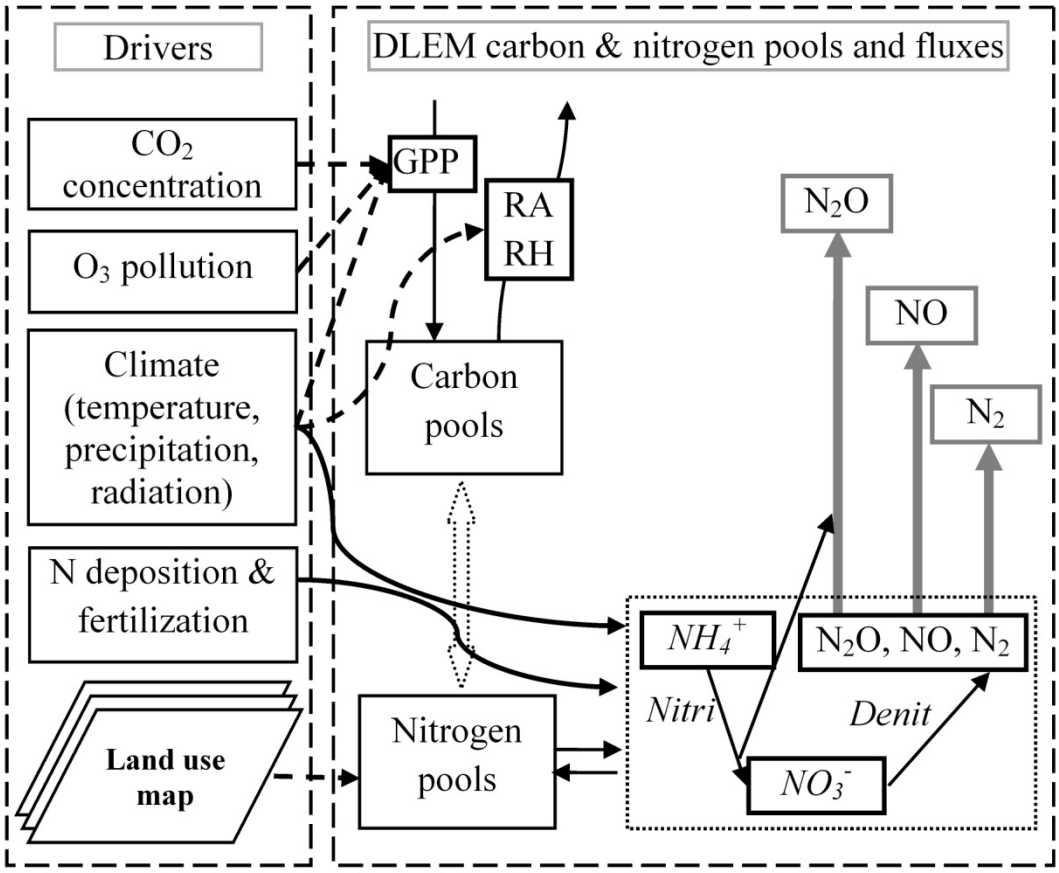
North America, as one of the extensively investigated continents, still lacks accurate estimate of N_2O emission due to limitations in method and observations. A number of studies estimated the N_2O emission by solely focusing on one ecosystem type or considering one global change factor (Del Grosso et al., 2006; Xu et al., 2008; Potter et al., 1996). For example, Li et al estimated N_2O emission from cropland in the United State of America (USA) by using a process-based model DNDC (Li et al., 1996). Del Grosso et al estimated the N_2O flux from cropland in the USA by using the DAYCENT model (Del Grosso et al., 2006). One of our previous studies simulated the regional flux of N_2O over North America during 1979-2008 by using a process-based ecosystem model, driven by multiple global change factors including climate variability,

elevated atmospheric CO_2, N deposition, O_3 pollution, N fertilization, and land use change (Tian et al., 2009b). In this study, we will further our analysis and attribute the variations in terrestrial N_2O at spatial and temporal scales to various factors.

Specifically, the objectives of this study are 1) to examine the factorial contributions to the spatial variation of terrestrial N_2O flux over North America during 1979-2008; 2) to quantify the factorial contributions to the temporal variations in terrestrial N_2O flux over North America during 1979-2008; 3) to quantify the factorial contributions to the 30-year accumulative flux of N_2O over North America at both continental and country levels; 4) to identify the major factors responsible for the spatial and temporal variations in terrestrial N_2O flux at both continental and country levels. The global change factors that will be evaluated in this study include climate variability, rising 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 N_2O flux contributed from all factors together by the changes in N_2O flux caused by six individual factors (see Experiment design section for the detail information).

2. Materials and methods

2.1. Brief description of the model used in this study



Major processes: *Ntri*: Nitrification; *Denit*: Denitrification; GPP is the gross primary productivity; RA is the autotrophic respiration from plant, and RH is the heterotrophic respiration; 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 N_2O processes.

Fig. 20 Conceptual diagram showing major processes for N_2O flux in response to multiple global change factors in the DLEM model

The model used in this study is an integrative ecosystem model DLEM which couples major biogeochemical cycles, hydrological cycles, and vegetation dynamics to make daily, spatially-explicit estimates of carbon (C), N, and water fluxes and pool sizes in terrestrial ecosystems. 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 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 forest, grassland, shrub, tundra, desert, wetland, and cropland. The simulated results have been compared with independent field data and satellite products. The DLEM operates at a daily time step and at varied spatial resolutions, from meters to kilometers, from regional to global. The detailed information for DLEM could be referred to our previous publications (Liu et al., 2008; Lu, 2009; Ren, 2009; Ren et al., 2007a; Ren et al., 2007b; Tian et al., 2010a, 2010b; Zhang, 2008; Zhang et al., 2007a; Xu et al., 2010) and the N_2O module has been described in detail in Tian et al. (Tian et al., 2010b).

In the DLEM, both denitrification and nitrification processes are simulated as one-step process as we do not consider the mid-products in each process. Nitrification, a process converting ammonium into nitrate, is simulated as a function of soil temperature, moisture, and the NH_4^+ concentration (Lin et al., 2000; Tian et al., 2009c). Denitrification, through which the nitrate is converted into N gases, is simulated in the DLEM as a function of soil temperature, moisture, and the NO_3^- concentration (Lin et al., 2000; Tian et al., 2009b). All the products of nitrification and denitrification that leave system are N-containing gases. The empirical equation

reported by Davidson et al (Davidson et al., 2000) is used to separate N_2O from other gases (mainly NO and N_2).

In summary, multiple global change factors yield direct and/or indirect impacts on N_2O processes in the DLEM (Fig. 20), which could be expressed as the following equation.

$$\boldsymbol{N_2O = V_{max} f(C_a, w_i, T_{air}, APAR) f(ozone) f(N)} \qquad \text{Equation 25}$$

where N_2O is the N_2O flux; V_{max} is the maximum rate of N_2O production via nitrification and denitrification; $f(C_a, w_i, T_{air}, APAR)$ describes the effects of air temperature, atmospheric CO_2 concentration, soil moisture, daily temperature, and absorbed photosynthetically active radiation on N_2O flux through its photosynthesis; $f(O_3)$ describes the effects of O_3 pollution; $f(N)$ describes the effect of N input; C_a is atmospheric CO_2 concentration, w_i is soil moisture; T_{air} is daily temperature, $APAR$ is absorbed photosynthetically active radiation. It should be noted that the above equation solely includes the direct and indirect effects from multiple global change factors on N_2O flux; the other environmental factors which might influence N_2O flux were not incorporated in the equation, for example, soil pH, soil porosity.

2.2. Study area and input data

This section is as same as the content in section 2.2 in the Chapter 3.

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 N_2O flux over North America, we conducted nineteen simulations in this study (Table 10; same as that in Chapter 3). One overall simulation was set up to simulate the terrestrial N_2O 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 N_2O 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. 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 10).

The implementation of DLEM simulation includes the following steps: 1) equilibrium run, 2) spinning-up run and 3) transient run. In this study, we used potential vegetation, long-term mean climate during 1979-2008, the concentration levels of N deposition, O_3, atmospheric

CO_2 in the year 1900 to drive the model run to an equilibrium state (i.e. the inter-annual variations are < 0.1 g m^{-2} for carbon storage, < 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 the terrestrial N_2O flux. Only the outputs between 1979 and 2008 were analyzed to show the spatial and temporal patterns of terrestrial N_2O flux in North America's terrestrial ecosystems. Urban was treated as grassland, which is as same as the strategy in other terrestrial biosphere model (McGuire et al., 2001). Baseline flux was defined as the N_2O 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

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

2.5. Statistic

The regression analysis was used in this study to find the long-term changing trend of input data and N_2O fluxes generated by various simulations. All the statistic 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

This section is as same as the content in section 3.1 in the Chapter 3.

3.2. Spatial distribution of N_2O flux over North America during 1979-2008

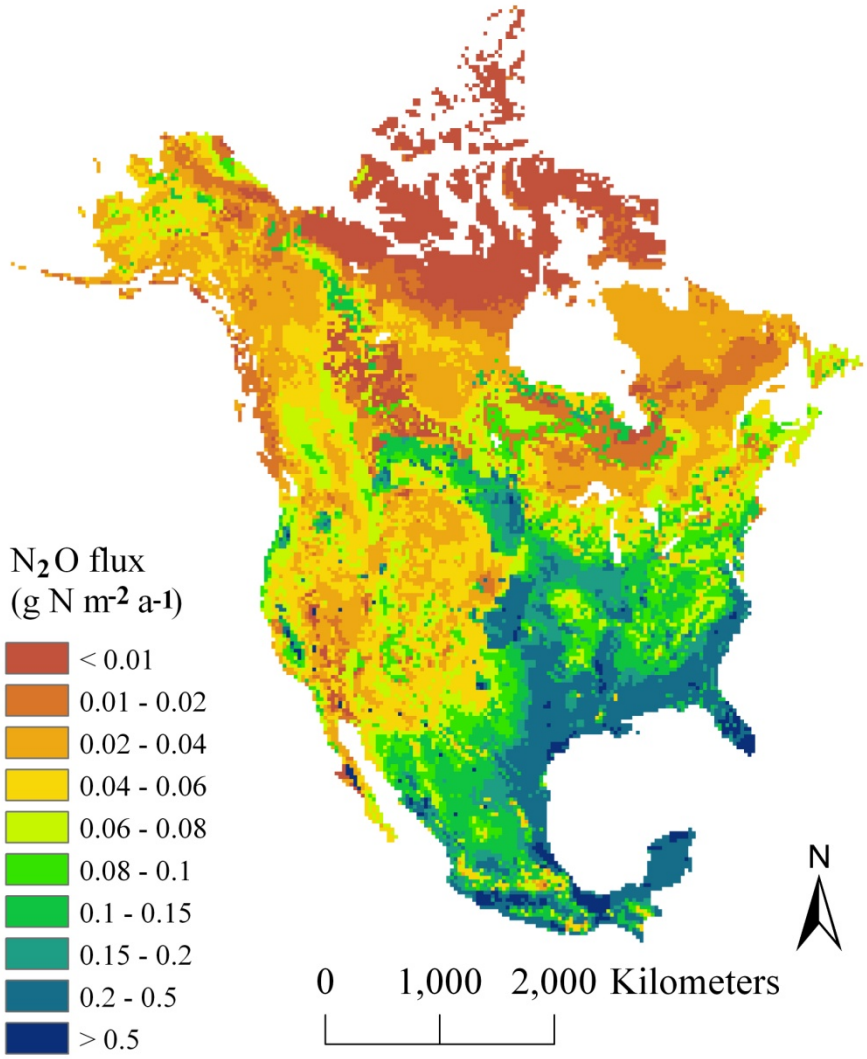


Fig. 21 Spatial variations of terrestrial N_2O fluxes caused by global change factors over North America from 1979 to 2008

The terrestrial N_2O flux over North America showed a significant spatial variation, with a strong source in southeastern continental North America including southeastern USA and majority of Mexico, and a weak source in northern part of North America (Fig. 21). The southern USA features high N_2O emission, up to approximately 1 g N m^{-2} a^{-1}, while the western USA has relatively low N_2O emission rate; there was a north-to-south increasing gradient of N_2O emission across Canada; the N_2O emission rate was as low as 0.0001 g N m^{-2} a^{-1} in northern Canada, and as high as approximately 0.4 g N m^{-2} a^{-1} in southern Canada; the entire Mexico acted as a strong source for N_2O, with the national N_2O emission being 0.21 ± 0.03 g N m^{-2} a^{-1} over the past 30 years.

3.3. Spatial contribution to the spatial variations in temporal N_2O flux during 1979-2008

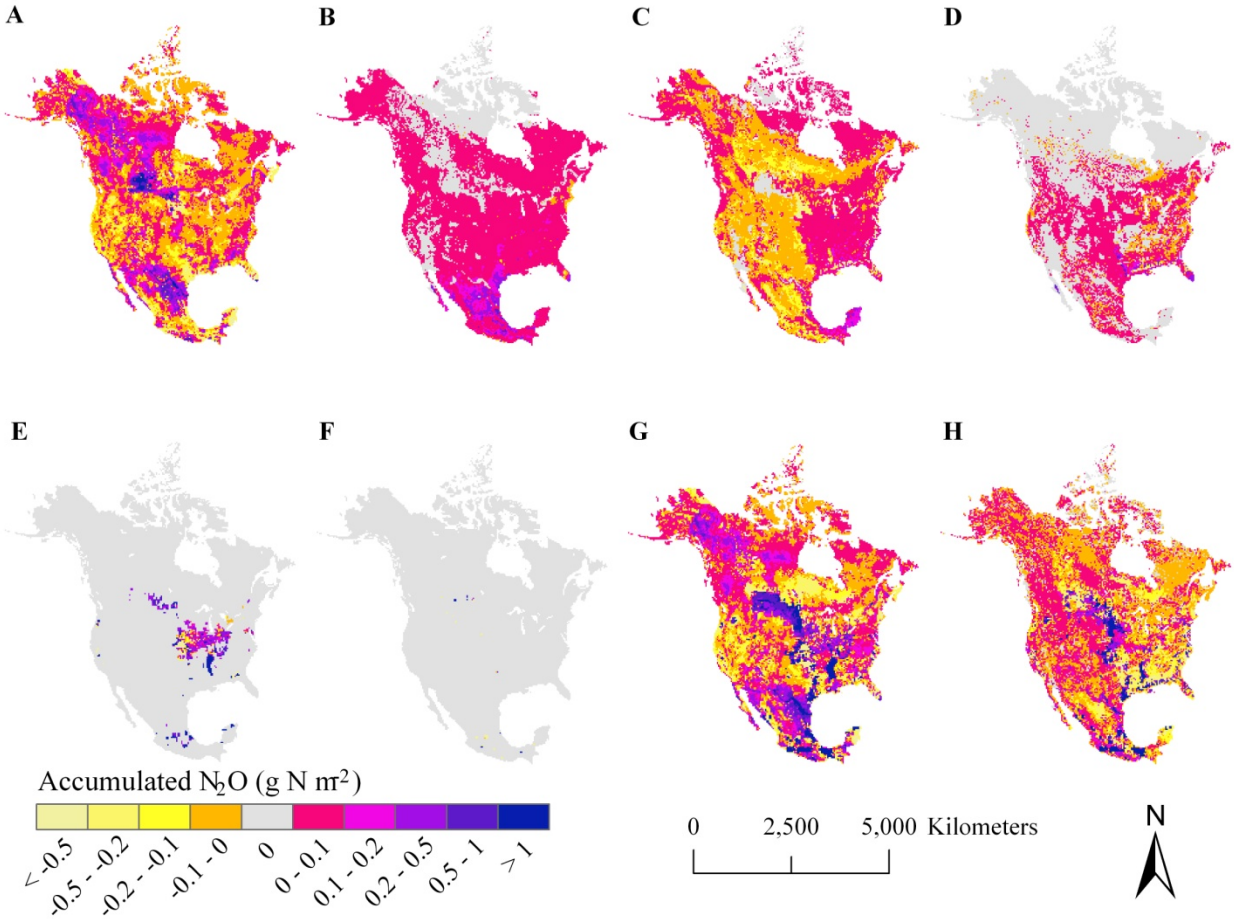


Fig. 22 Factorial contributions to the spatial variations in accumulated N_2O flux over North America from 1979 to 2008 (A: climatic variability; B: N deposition; C: CO_2; D: O_3 pollution; E: N fertilization; F: land conversion; G: all combined; H: interaction)

For a N-balanced system, a portion of N will leave system as N_2O gas; when external factors were posed on, the terrestrial N_2O flux will be changed; the terrestrial N_2O flux at N-balanced state is background emission, the changes in terrestrial N_2O flux caused by external factors is external-factor-induced N_2O flux. Throughout the study period, the accumulative N_2O emission over North America was composed of two components; one is the baseline emission as the background emission through study period; the other component is the flux contributed from changes in various individual and interactive effects of several global change factors. After removing the baseline flux of N_2O, the left flux of N_2O was contributed from six global change factors and their interaction.

Over the past 30 years, climate variations enhanced N_2O emission over majority of the continental North America, while decreased N_2O emission over portions of western Alaska, eastern USA and eastern Canada (Fig 22A); N deposition enhanced N_2O emission over the most of continental North America with prominent increases over the Mexico (Fig 22B); elevated atmospheric CO_2 enhanced N_2O emission in eastern USA and northeastern Canada, while decreased N_2O emission in western USA, western Canada and majority of Mexico (Fig 22C); O_3 pollution increased N_2O emission in majority of USA and southern Canada, while did not yield significant impacts on N_2O flux in majority of Canada (Fig 22D); N fertilization enhanced N_2O emission in central USA which is agricultural land (Fig 22E), while land use conversion only affects a small amount of area which experienced land use conversion between natural vegetation and cropland or urban in the past few years (Fig 22F); interactive effect among global change factors enhanced N_2O emission across portions of North America including northwestern

continental North America, while decreased N_2O emission in eastern USA and Canada (Fig 22G); combining all the effects from various global change factors, the N_2O emission was enhanced across majority of continental North America, yet was decreased in portions of continental North America (Fig 22H).

3.4. Temporal patterns of N_2O flux over North America during 1979-2008

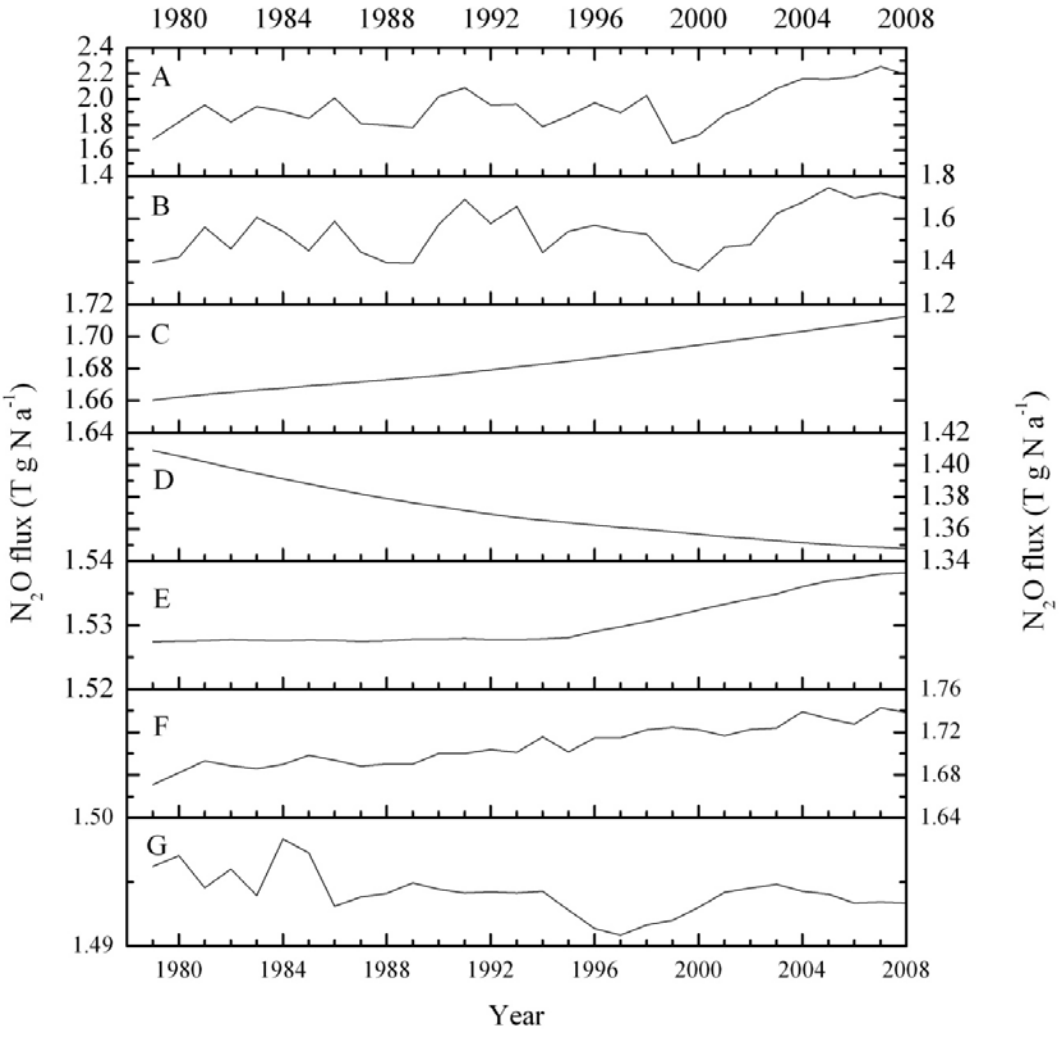


Fig. 23 Temporal variations of terrestrial N_2O flux caused by global change factors over North America from 1979 to 2008 (A: all combined simulation; B: climate only simulation; C: N deposition only simulation; D: CO_2 only simulation; E: O_3 pollution only simulation; F: N fertilization only simulation; G: land conversion only simulation)

The continental-level terrestrial flux of N_2O over the North America showed a significant inter-annual fluctuation during 1979-2008 (Fig 23). The highest N_2O emission was 2.25 T g N_2O-N a^{-1} in 2007, and the lowest was 1.66 T g N_2O-N a-1 in 1999 (Fig 23). The mean annual N_2O flux over North America's terrestrial ecosystems was 1.94 ± 0.16 T g N_2O-N a^{-1}, with an overall increasing rate of 9.99 ± 2.76 G g (1G g = 10^9 g) N_2O-N a^{-1}, over the past 30 years. The long-term increasing trend and inter-annual fluctuation in terrestrial N_2O flux were contributed from multiple global change factors (Fig 23). Climate variability exerted a significantly inter-annual variation in N_2O flux, with an increasing rate of 6.10 ± 2.12 G g N_2O-N a^{-1} ($P = 0.008$). N deposition contributed to an increasing rate of 1.77 ± 0.03 G g N_2O-N a^{-1} ($P < 0.001$), and N fertilization contributed to an increasing rate at 2.06 ± 0.012 G g N_2O-N a^{-1} for continental N_2O emission ($P < 0.001$). The continuous increased atmospheric CO_2 concentration continuously decreased N_2O emission which generated a decreasing rate of 2.06 ± 0.08 G g N_2O-N a^{-1} ($P < 0.001$) over the study period, O_3 pollution made a positive effect on N_2O emission at a rate of 0.38 ± 0.04 G g N_2O-N a^{-1} ($P < 0.001$), while land conversion yielded decreasing trend at 0.10 ± 0.03 G g N_2O-N a^{-1} ($P = 0.0025$), respectively.

3.5. Factorial contribution to the accumulated N_2O flux over North America during 1979-2008

We summed up the individual global change factor-induced changes in N_2O flux over 30 years to analyze the contributions of six single factors and their interaction (Fig. 24). To express the uncertainties associated with the accumulated N_2O 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. During the 30-year study period, the accumulative N_2O flux over North America was 58.17 ± 0.85 T g N_2O-N, of which 55.35 ± 0.56 T g N_2O-N was contributed from baseline

flux and 2.81 ± 0.98 T g N_2O-N was caused by global change factors. Elevated atmospheric CO_2 decreased the N_2O emission by 0.51 ± 0.07 T g N_2O-N from North America's terrestrial ecosystems, while all the other single factors increased N_2O emission. Climate variations, N deposition, O_3 pollution, N fertilization, land conversion, and multiple-fact interaction enhanced continental N_2O emission by 0.56 ± 0.68, 0.50 ± 0.07, 0.10 ± 0.02, 0.92 ± 0.09, 0.16 ± 0.01, and 1.10 ± 0.37 T g N_2O-N, respectively (Table 16).

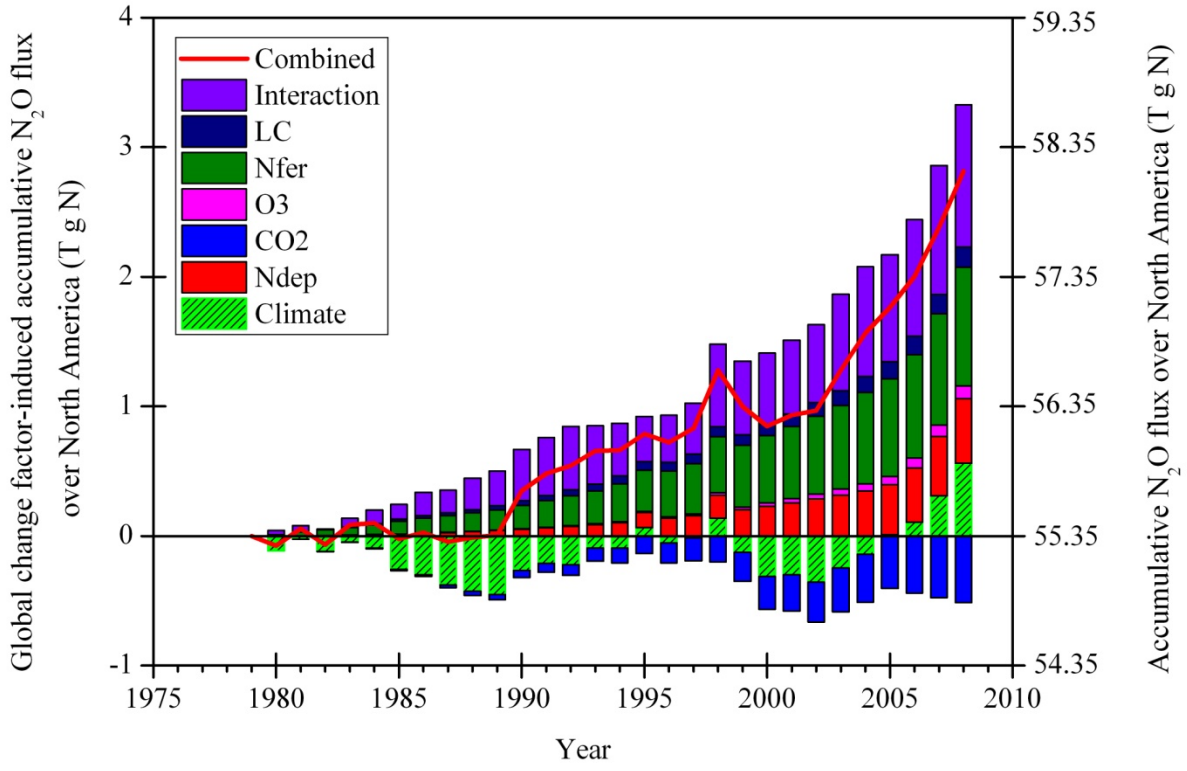


Fig. 24 Factorial contributions to accumulated N_2O flux over North America during 1979-2008 (The right Y-axis shows the accumulative N_2O flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilization; *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)

Table 16 Factorial contributions to the accumulated N_2O from 1979 to 2008

		Baseline	Climate	Ndep	CO_2	O_3	Nfer	Land conversion	Interaction	Total flux
USA	Accumulative N_2O flux (T g N)	31.81 ± 0.42	-0.10 ± 0.47	0.22 ± 0.03	-0.25 ± 0.04	0.08 ± 0.02	0.55 ± 0.07	0.18 ± 0.02	0.18 ± 0.26	32.68 ± 0.45
	Percentage (%)	97.34	-0.31	0.67	-0.76	0.25	1.68	0.56	0.56	100
Canada	Accumulative N_2O flux (T g N)	10.23 ± 0.12	0.55 ± 0.29	0.03 ± 0.005	-0.20 ± 0.02	0.004 ± 0.001	0.09 ± 0.01	0.001 ± 0.002	-0.10 ± 0.08	10.60 ± 0.22
	Percentage (%)	96.47	5.18	0.27	-1.89	0.04	0.83	0.01	-0.91	100
Mexico	Accumulative N_2O flux (T g N)	13.31 ± 0.38	0.12 ± 0.42	0.25 ± 0.04	-0.07 ± 0.01	0.01 ± 0.003	0.28 ± 0.02	-0.03 ± 0.01	1.01 ± 0.18	14.88 ± 0.46
	Percentage (%)	89.43	0.78	1.68	-0.44	0.08	1.89	-0.19	6.78	100
NA	Accumulative N_2O flux (T g N)	55.35 ± 0.56	0.56 ± 0.68	0.50 ± 0.07	-0.51 ± 0.07	0.10 ± 0.02	0.92 ± 0.09	0.16 ± 0.01	1.10 ± 0.37	58.17 ± 0.85
	Percentage (%)	95.16	0.97	0.86	-0.88	0.17	1.58	0.27	1.89	100

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; *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 fertilization; *Land conversion* represents the impacts of land cover change only; *Interaction* represents the balance of all interactive effects of the six environmental factors

Table 17 Contributions from individual climate variable to the climate-induced N_2O accumulation (T g N) from 1979 to 2008

	Precipitation	Relative humidity	Solar radiation	Temperature	Interaction	Total flux
US	-2.62 ± 0.39	-3.71 ± 0.35	-3.64 ± 0.35	-0.37 ± 0.42	-10.25 ± 1.05	-0.10 ± 0.47
Canada	0.07 ± 0.25	-1.73 ± 0.16	-1.80 ± 0.16	-1.39 ± 0.17	5.40 ± 0.49	0.55 ± 0.29
Mexico	-0.18 ± 0.44	-0.78 ± 0.31	-0.79 ± 0.31	0.56 ± 0.34	1.31 ± 0.92	0.12 ± 0.42
North America	-2.74 ± 0.62	-6.22 ± 0.50	-6.23 ± 0.51	-1.21 ± 0.59	16.96 ± 1.49	0.56 ± 0.68

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

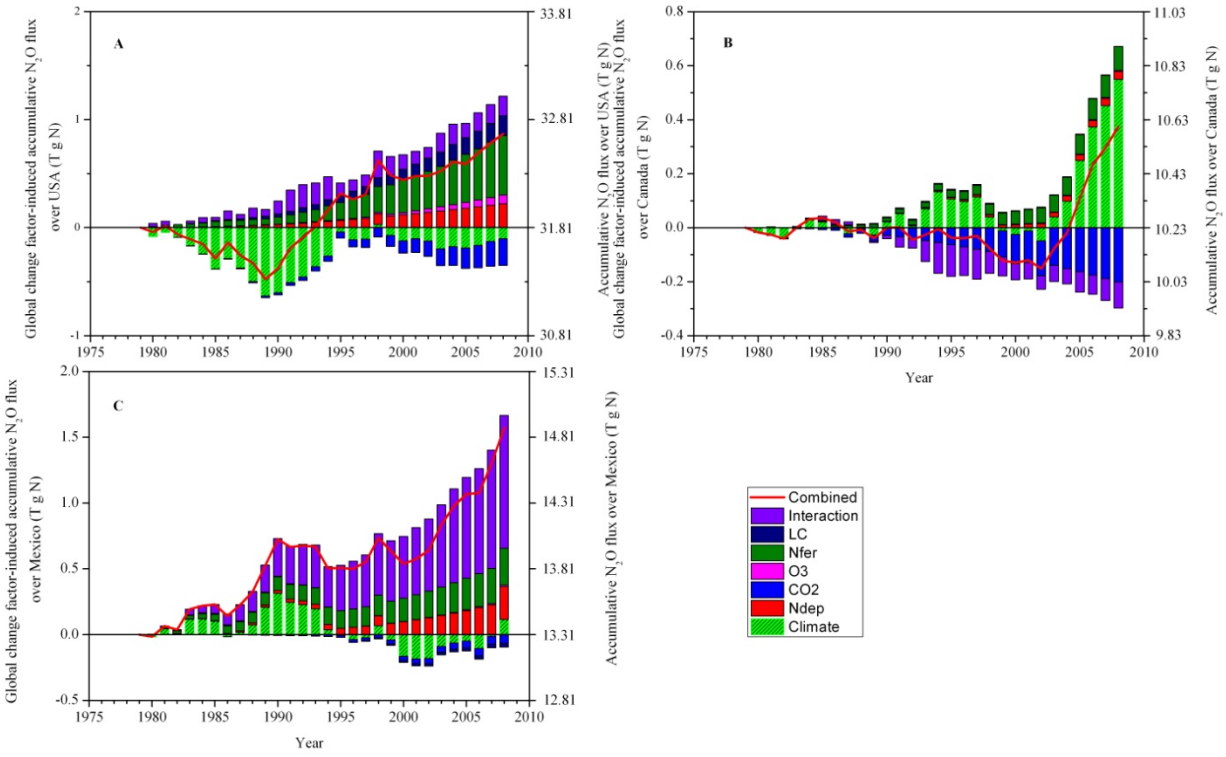


Fig. 25 Factorial contributions to accumulated N_2O flux at country-level during 1979-2008 (A: United States of America; B: Canada; C: Mexico) (The right Y-axis shows the accumulative N_2O flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilization; 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)

To examine the factorial contributions to accumulated terrestrial N_2O flux for three countries, we further partitioned continental level N_2O flux into country-level fluxes (Fig. 25). The 30-year accumulative N_2O emission was 32.68 ± 0.45 T g N_2O-N for USA, 10.60 ± 0.22 T g N_2O-N for Canada, and 14.88 ± 0.0.46 T g N_2O-N for Mexico, respectively (Table 16). For USA, climate variability and elevated atmospheric CO_2 decreased N_2O emission by 0.10 ± 0.47 T g N_2O-N and 0.25 ± 0.04 T g N_2O-N during the past 30 years, while N deposition, O_3 pollution, N fertilization, land conversion, and multiple-factor interaction increased the country-level N_2O emission (Table 16). For Canada, climate variations, N deposition, O_3 pollution, and N fertilization increased N_2O emission by 0.55 ± 0.29, 0.03 ± 0.005, 0.004 ± 0.001, and 0.09 ± 0.01

T g N_2O-N, respectively, during the 1979-2008; while elevated atmospheric CO_2 and multiple-factor interaction decreased N_2O emission by 0.20 ± 0.02 T g N_2O-N and 0.10 ± 0.08 T g N_2O-N, respectively (Table 16). For Mexico, climate variability, N deposition, O_3 pollution, N fertilization, and multiple-factor interaction enhanced N_2O emission by 0.12 ± 0.42, 0.25 ± 0.04, 0.01 ± 0.003, 0.28 ± 0.02, and 1.01 ± 0.18 T g N_2O-N, respectively, during the time period of 1979-2008; while elevated atmospheric CO_2 and land conversion decreased N_2O emission by 0.07 ± 0.01 T g N_2O-N and 0.03 ± 0.01 T g N_2O-N, respectively (Table 16). All climate variables including precipitation, solar radiation, relative humidity, and temperature, made negative impacts on N_2O flux in North America and the US. While precipitation stimulated N_2O flux in Canada and temperature change enhanced N_2O emission from Mexico (Table 17). Overall, the global change factors enhanced N_2O emission at country- and continental levels over the study period.

For the accumulative N_2O fluxes, the baseline emission, climate variability, N deposition, elevated atmospheric CO_2, O_3 pollution, N fertilization, land conversion, and multiple-factor interaction contributed 95.16%, 0.97%, 0.86%, -0.88%, 0.17%, 1.58%, 0.27%, and 1.89%, respectively, to the continental-level emission from the entire North America. For the country-level accumulative terrestrial N_2O fluxes, the baseline emission, climate variability, N deposition, elevated atmospheric CO_2, O_3 pollution, N fertilization, land conversion, and interaction contributed 97.34%, -0.31%, 0.67%, -0.76%, 0.25%, 1.68%, 0.56%, and 0.56%, respectively, to the country-level emission from USA, 96.47%, 5.18%, 0.27%, -1.89%, 0.04%, 0.83%, 0.01%, and -0.91%, respectively, to the country-level emission from Canada, and 89.43%, 0.78%, 1.68%, -0.44%, 0.08%, 1.89%, -0.19%, and 6.78%, respectively, to the country-level emission from Mexico (Table 16).

3.6. Factorial contribution to the inter-annual variations in N_2O flux over North America

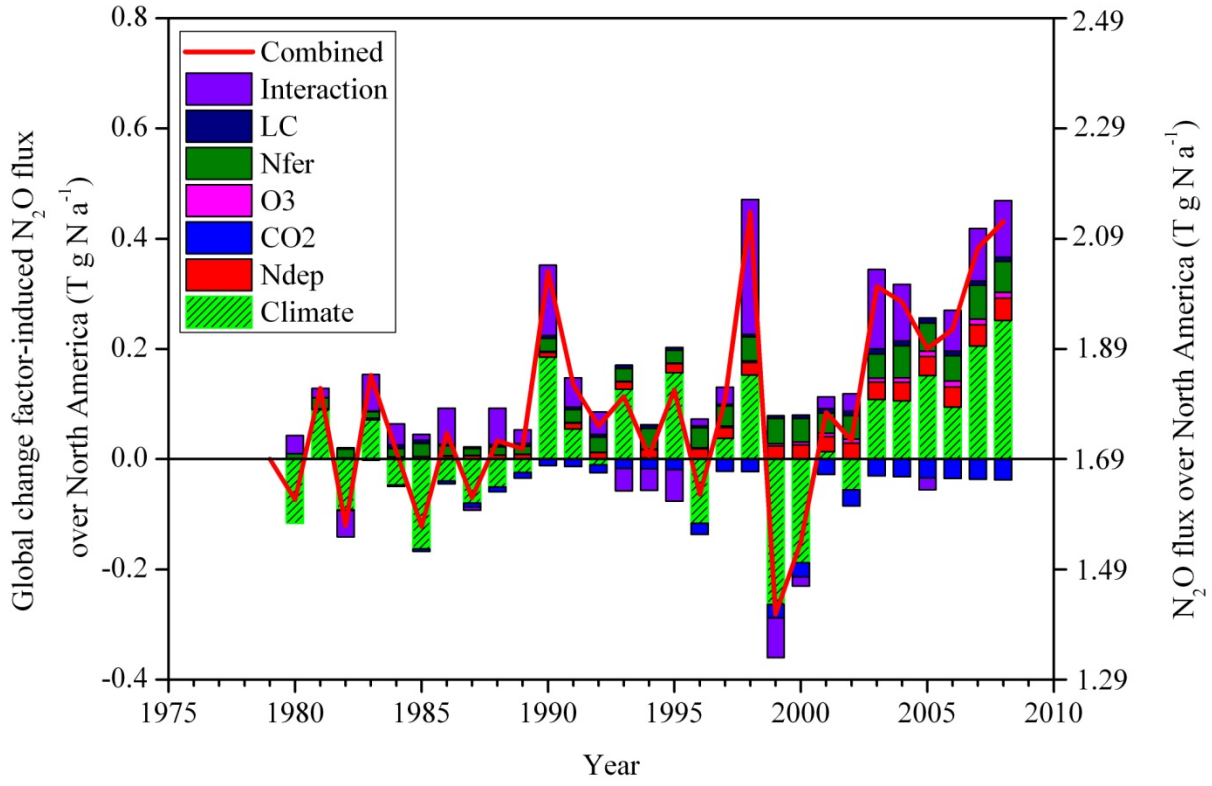


Fig. 26 Factorial contribution to the inter-annual variations in N_2O flux over North America (The right Y-axis shows the accumulative N_2O flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilization; *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)

Inter-annual variation is one of major attributes of ecosystem processes; it may be caused by internal mechanisms or external environmental control. Inter-annual variation in terrestrial N_2O flux has been shown over North America from 1979 to 2008 (Fig 23). To examine the controlling factors for this inter-annual variation, we further attributed the changes in terrestrial N_2O flux for each year to six global change factors and their interaction; and the results were shown in Fig 26. Over the entire North America, rising atmospheric CO_2 continuously decreased, while N deposition and N fertilization continuously increased terrestrial N_2O emission. O_3

pollution yield very small positive effects on terrestrial N_2O emission, while land conversion yielded small yet fluctuated effects on terrestrial N_2O emission (Fig 26). The climate variability primarily dominated the inter-annual fluctuation in terrestrial N_2O flux from 1979 to 2008. Climate variability and multiple-factor interaction co-dominated the increases in N_2O emission in three specific time periods: 1979-1983, 1989-1993, and 2001-2008. For the specific years, such as 1992, 1994, and 2001 when the climatic contribution to the N_2O flux is neutral, the interactive effect among multiple factors and other factors dominated the changes of terrestrial N_2O flux comparing to baseline flux (Fig. 26).

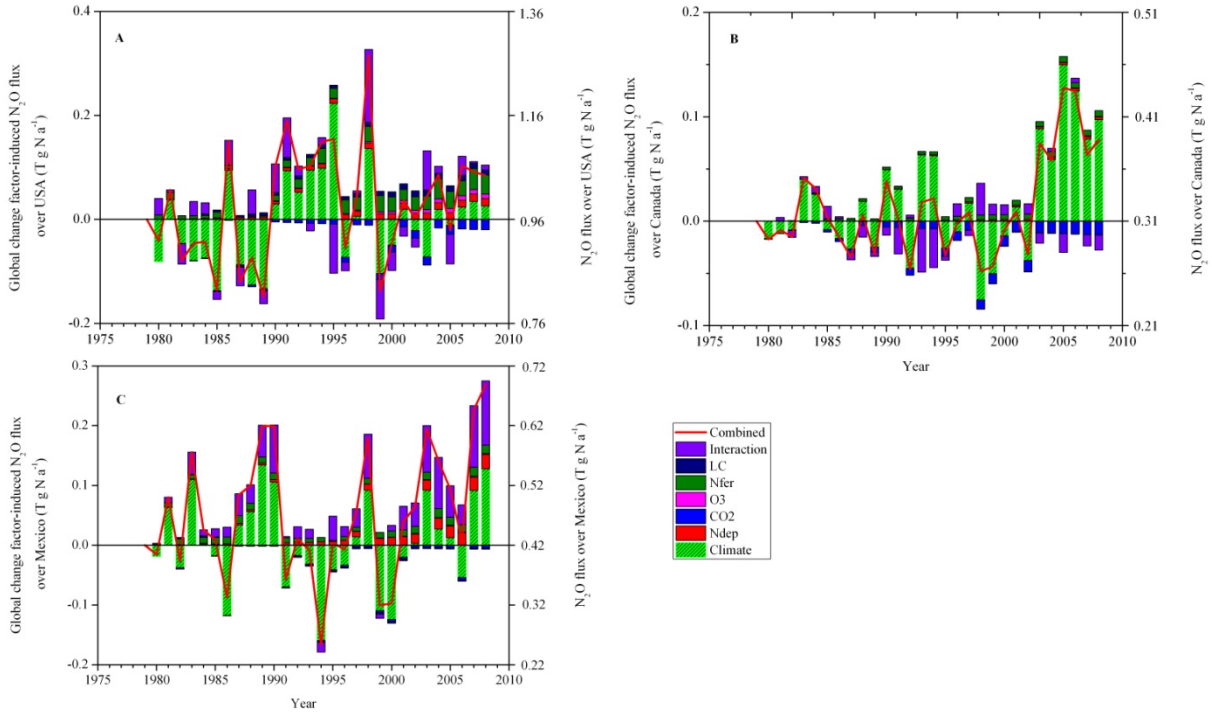


Fig. 27 Factorial contribution to the inter-annual variations in N_2O flux by country (A: United States of America; B: Canada; C: Mexico) (The right Y-axis shows the accumulative N_2O flux with baseline; *Interaction* means contribution from multiple-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilization; 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)

After partitioning continental flux into country-level emissions of N_2O, we further analyzed and identified the major factors controlling the inter-annual fluctuation in terrestrial N_2O over each country. It is found that the major factors leading to inter-annual fluctuation in terrestrial N_2O flux varied over different countries. Climate variability and multiple-factor interaction c-dominated the inter-annual fluctuations in terrestrial N_2O flux over USA. During the period of 1979-1988 and 1990-1996 the climatic effects dominated the terrestrial N_2O emission over USA; meanwhile, the interactive effects among factors contributed to the flux of terrestrial N_2O over entire time period (Fig. 27A).

Climate variability outweighed other factors in controlling the increases in terrestrial N_2O emission over Canada during most of the study period (Fig. 27B). For instance, the climate-induced increases in terrestrial N_2O flux are much higher than contributions from other factors over the time period of 2003-2008. However, in the years of 1993-1994, the climate-induced increases in terrestrial N_2O were partially offset by other factors' effects. Similar to USA, the rising atmospheric CO_2 continuously decrease N_2O emission. O_3 pollution, N input and land conversion exerted minor effects on terrestrial N_2O flux. The multiple-factor interaction yielded very complex effects; it varied significantly through the study period, positive or negative, small or large. For Mexico, climate variability and multiple-factor interaction played a predominant role in controlling inter-annual variations in terrestrial N_2O flux, the contributions from all other factors were in small magnitude (Fig. 27C).

4. Discussion

4.1. Comparison with other studies

N_2O flux was primarily controlled by environmental factors, substrate availability, and energy source (Brumme et al., 1999; Williams et al., 1992; Conrad, 1996). Global change

factors alter terrestrial N_2O flux through their effects on these processes (Dong et al., 2003; Kettunen et al., 2005; Mcswiney and Robertson, 2005; Kanerva et al., 2007; Zhang et al., 2007b). As consistent with previous studies (Mosier et al., 1991; Li et al., 1996; Mcswiney and Robertson, 2005; Zhang et al., 2007b), N input including N deposition and N fertilization enhanced the N_2O emission from terrestrial ecosystems. As the theory of progressive N limitation predicts (Luo et al., 2004), rising atmospheric CO_2 could lead to low N availability in soil and thus lead to low N_2O emission (Kettunen et al., 2005; Barnard et al., 2004). In this study, the elevated atmospheric CO_2 substantially decreased the N_2O emission from terrestrial ecosystem over North America, which might due to the N limitation for major biomes throughout the entire North America (Vitousek and Farrington, 1997; Aber and Melillo, 2001). O_3 has been confirmed as a pollutant which may decrease productivity (Wang et al., 2007), and thus carbon source for microbial processes responsible for N_2O production, and finally cause decrease in N_2O emission. However, it is also reasonable to infer that the O_3 pollution inhibits productivity (Wang et al., 2007; Ren, 2009); and less N uptake might increase N availability in soil and in turn increase N_2O emission. Field experiments are lacking these two hypotheses. This study confirmed that the O_3 pollution enhanced N_2O emission from terrestrial ecosystems.

We also compared out modeled results against previous studies to verify the factorial effects on N_2O flux for major biomes (Table 18). Our estimated continental-level average response to double CO_2 is -36.33% for forest ecosystems, which is consistent with *Phillips et al's* report that the N_2O emission will be suppressed by elevated CO_2 (2001). DLEM simulated a decrease of 17.54% of N_2O emission from grassland which is consistent with *Baggs et al's* study (2003). All field observations reported that the effects of elevated CO_2 on N_2O emission depend on N availability; this could be explained by N limitation theory (Luo et al., 2004). Several

studies have concluded that majorities of forest and grassland on the planet are N limited (Aber et al., 1998; Aber and Melillo, 2001; Vitousek and Farrington, 1997; Vitousek et al., 1997), so the elevated CO_2 might suppress N_2O emission from these ecosystems. Our study found that the elevated CO_2 suppressed N_2O emission over North America's forest and grassland. One study reported that the elevated atmospheric CO_2 might stimulate N_2O emission during growing season when mineral N is limited; the authors attributed this phenomenon to fungal activity (Kammann et al., 2008). This calls for further study of incorporation of soil biota in large-scale ecosystem models.

This study also found that N deposition could stimulate N_2O emission in all major biomes, which is consistent with field observations (Liu and Greaver, 2009; Ambus and Robertson, 2006; Kettunen et al., 2005). DLEM simulated the N deposition could stimulate N_2O emission at rates of 28.55 ± 0.24 mg N m^{-2} a^{-1} / g N m^{-2} a^{-1} for forest, 49.15 ± 0.19 mg N m^{-2} a^{-1} / g N m^{-2} a^{-1} for grassland, 47.77 ± 0.88 mg N m^{-2} a^{-1} / g N m^{-2} a^{-1} for wetland, and 22.39 ± 0.37 mg N m^{-2} a^{-1} / g N m^{-2} a^{-1} for cropland, respectively. These responses are consistent with Pilegaard's summarized results that the emission of N in N-oxides was 2-32% of the amount of N in atmospheric input to the soil (Pilegaard et al., 2006), while are larger than the summarized values by *Liu and Greaver* (2009). This might due to the lack of data collection in Liu and Greaver (2009) or others, which worth more investigation. There is only one reported field study for the effect of O_3 pollution on N_2O emission from grassland (Kanerva et al., 2007). The DLEM-estimated result is different from filed observation in the reported field study, yet both reported responses are very small, nearly neutral effects. O_3 has been found as an inhibitor of plant growth (Kanerva et al., 2007;Wang et al., 2007), worse plant growth means lower N uptake which will increase N availability in soil, and thus stimulate N_2O emission.

Table 18 Comparison of factorial effects on N_2O fluxes against other studies (positive values mean increase; negative values mean decrease in N_2O emission)

	Biome	Experiment design	Simulated	Others	Literature
Elevated CO_2	Forest	Double CO_2	-36.33%	Negative, neutral or positive effects, depending on seasons and nitrogen availability	Ambus and Robertson, 1999;Phillips et al., 2001
	Grassland	600 ppm *vs* 360ppm CO_2	-17.54%	-5.65% ~ -26.01% for low nitrogen input; 1.43% ~ 62.27% for high nitrogen input	Baggs et al., 2003;Ineson et al., 1998;Kanerva et al., 2007;Ambus and Robertson, 2006
Nitrogen deposition	Forest		28.55 ± 0.24 (mg N m^2 a^{-1} / g N m^2 a^{-1})	6 ± 1 (mg N m^2 a^{-1} / g N m^2 a^{-1})	Liu and Greaver, 2009
	Grassland		49.15 ± 0.19 (mg N m^2 a^{-1} / g N m^2 a^{-1})	6 ± 1 (mg N m^2 a^{-1} / g N m^2 a^{-1})	Liu and Greaver, 2009
	Wetland		47.77 ± 0.88 (mg N m^2 a^{-1} / g N m^2 a^{-1})	36 ± 13 (mg N m^2 a^{-1} / g N m^2 a^{-1})	Liu and Greaver, 2009
	Cropland		22.39 ± 0.37 (mg N m^2 a^{-1} / g N m^2 a^{-1}) for nitrogen deposition; 16.35 ± 0.45 (mg N m^2 a^{-1} / g N m^2 a^{-1}) for nitrogen fertilization	9 ± 1 (mg N m^2 a^{-1} / g N m^2 a^{-1})	Liu and Greaver, 2009
O_3 pollution	Grassland	40-50 ppb in Open-top chambers	1.5%	Decrease yet not significantly	Kanerva et al., 2007

4.2. Spatial pattern of N_2O flux and controlling factors

Spatial variations in terrestrial N_2O flux over North America simulated in this study were consistent with *Potter et al's* (Potter et al., 1996) study and *Xu et al's* study (Xu et al., 2008). The major source for atmospheric N_2O locates in southeastern continental North America including south part of central Canada, southeastern USA and entire Mexico, because the cropland in central USA and the majority of tropical rain forest in southeaster USA and entire Mexico (Fig 21). The weak source in northern Canada is probably due to its low temperature and rainfall as N_2O emission is significantly control by temperature and soil moisture (Chapuis-Lardy et al., 2007; Conrad, 1996; Goldberg and Gebauer, 2009).

4.3. Temporal pattern of N_2O flux and controlling factors

There is a substantial inter-annual fluctuation in terrestrial N_2O flux over North America during 1979-2008, which is possible due to complicated impacts from multiple factors (Chapuis-Lardy et al., 2007; Conrad, 1996; Ambus and Robertson, 1999). The highly varied climatic variability dominated the inter-annual flux of N_2O over North America, and solely dominated the N_2O flux over Canada. N deposition, O_3 pollution, atmospheric CO_2 concentration increased at relatively stable rates through the past 30 years, which resulted in the long-term increasing trend of N_2O flux; and the significantly inter-annual variations in climatic variables lead to the inter-annual variations of N_2O flux (Fig 27).

Using stepwise regression analysis, we found that the climate variability is the predominating factor controlling inter-annual variations in terrestrial N_2O flux at both continental and country levels. Following the climate variability, multiple-factor interaction played an essential role in contributing temporal variations in terrestrial N_2O flux. This is consistent with our previous analysis showing that the climate variability, followed by multiple-

factor interaction, co-dominated inter-annual variations in terrestrial N_2O flux at both continental and country levels (sections 3.7 and 3.8 in this chapter).

4.4. Uncertainties

This study attributed the spatial and temporal variations in N_2O flux over North America's terrestrial ecosystems during 1979-2008, there are several issues need to be improved. First, because this study only considered the land conversion between cropland and natural vegetation, it will generate more accurate results if deforestation and afforestation are included. Second, the legacy effect of the previous 78 years (1901-1978) may cause some biases in this study which solely analyzed N_2O flux over the time period of 1979 – 2008; however, due to the issue of climate data availability we cannot do it in this study. This might need to be addressed in future research. Third, the soil may act as a sink of atmospheric N_2O (Chapuis-Lardy et al., 2007); however, owning to incomplete understanding of this phenomenon we did not incorporate the mechanisms in this study; so this study may overestimate the terrestrial N_2O flux at both continental and county levels. More field studies are needed to understand the mechanisms for N_2O sink in soil which will improve the regional estimation of N_2O flux. Fourth, although we have compared our estimated factorial impacts on N_2O flux, the effects of global change factors N_2O have not been comprehensively calibrated and validated because of the scarcity of field observations on factorial N_2O fluxes (Dermody, 2006). Last but not least, it will be an improvement if modeling study takes into account more environmental factors such as wild and prescribed fire, harvest, insects etc. For instance, one recent study reported the freeze-thaw could lead to an abrupt increase in N_2O emission from high latitudinal peat-land and lake (Pihlatie et al., 2009; Walter et al., 2006).

5. Conclusion

Factorial contributions to the N_2O flux over North America were examined at both continental and country levels by using a highly-integrated process-based ecosystem 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, this study is helpful in advancing our understanding of the dynamics of atmospheric N_2O concentration; it might also benefit the policy-making for curbing the increase in atmospheric N_2O concentration. The complicated effects of multiple-factor interaction on N_2O flux suggest that the current experiments which usually ignore the interactive effects from multiple-factor may lead to biases in the estimation of N_2O flux. This study also pointed out that the models driven by few global change factors may bring bias in estimating N_2O flux.

To our knowledge, this study is among the first effort to attribute the spatiotemporal variations in regional terrestrial N_2O flux to multiple global change factors over a long period of time. This study also provides insights for examination of the multiple-factor interactive effects on terrestrial N_2O flux. Given the advantages of modeling approach in quantification of N_2O flux, and the importance of field experiments in model improvement and flux estimation, a collaborative effort between field ecologists and modelers might be necessary for further investigation of the underlying mechanisms for spatial and temporal variations in N_2O flux.

Chapter 5. Local Sensitivity and Uncertainty Analyses for Terrestrial CH_4 and N_2O Fluxes over North America

Abstract

Process-based modeling approach is gaining popularity in regional estimation of CH_4 and N_2O fluxes. However, the uncertainties caused by parameters have not been widely evaluated at regional scale. In this study, we proposed an approach by combining local sensitivity analysis and local uncertainty analysis to evaluate the parameter-induced uncertainties in regional terrestrial fluxes of CH_4 and N_2O over North America as estimated by a process-based biogeochemistry model, dynamics land ecosystem model (DLEM). We first assumed that the parameters responsible for CH_4 and N_2O fluxes follow normal distribution. Then the local sensitivity analysis was conducted to identify the major parameters which are predominantly control the terrestrial fluxes of CH_4 and N_2O. Based on the explored distribution and the *priori* knowledge of all parameters, a local uncertainty analysis was further carried out to evaluate the uncertainties caused by parameters in the regional estimation of terrestrial CH_4 and N_2O fluxes over North America. The parameter, maximum rate of CH_4 production, followed by the parameters controlling atmospheric CH_4 oxidation, is the most sensitive parameter controlling the continental CH_4 flux. The major parameters controlling biome- and country-level CH_4 fluxes varied among countries and biomes; while the ubiquitous dominant parameter controlling biome-level, country-level, and continental N_2O fluxes is the maximum rate of denitrification. The

simulations concluded that the 95% confidence interval of terrestrial CH_4 flux over North America is (5.95 T g C a^{-1}, 23.10 T g C a^{-1}), and the 95% confidence interval of terrestrial N_2O flux is (0.75 T g N a^{-1}, 3.38 T g N a^{-1}). The spatial distributions of the uncertainties in terrestrial CH_4 and N_2O fluxes varied across the continental North America; the largest uncertainty in CH_4 flux locates in wetland, while the largest uncertainty in N_2O flux lies in tropical forest, followed by cropland. The level of uncertainties in regional estimates of CH_4 and N_2O are highly depending on the *priori* knowledge of parameters, which calls for more efforts in obtaining the accurate and precise value for each parameter in modeling approach.

1. Introduction

Process-based modeling approach has been broadly used in the regional estimation of terrestrial methane (CH_4) and nitrous oxide (N_2O) fluxes (Li et al., 2001; Potter et al., 2006; Potter et al., 1996; Tian et al., 2010b; Walter et al., 2001; Zhuang et al., 2007). However, the uncertainties associated with the estimates are rarely investigated (Potter, 1997; Potter et al., 1996; Walter et al., 2001). This is due to both the complexity of uncertainty sources (Haefner, 2005), namely input data, model structure, and parameters, and the challenges in the approach for uncertainty analysis (Tang and Zhuang, 2009; Varella et al., 2010; Werner et al., 2007).

In the ecological studies, the uncertainties caused by input data could be addressed by the improvement in data development (Hakanson, 2003; Loveland et al., 2000; Sims et al., 2008), and/or the comparison of multiple-source input data (Fritz and See, 2008; Haefner, 2005); the uncertainties involved in model structure might be evaluated by systematic analysis (Hakanson, 2003; Hosack et al., 2008) and be addressed by integration of advanced mechanisms (Haefner, 2005; Hakanson, 2003; Yang et al., 2009). The parameter-induced uncertainties, however, are

complicated and challenging to be addressed (Hakanson, 2003; van Bodegom et al., 2000) because it involves not only considerable modeling effort, but also lots of mathematical and statistic knowledge (Cacuci, 2003; Haefner, 2005; Prihodko et al., 2008; Tatang et al., 1997). A number of studies were conducted at site level to evaluate the uncertainties in ecosystem processes caused by parameterization processes (Tang and Zhuang, 2009; Varella et al., 2010), only few studies were conducted to evaluate the parameter-induced uncertainties at regional scale (Li et al., 2004; Werner et al., 2007).

Global sensitivity and uncertainty analysis has been used for identification of the major parameters (Tang and Zhuang, 2009), and evaluation of the uncertainties involved in parameters (Li et al., 2004; van Bodegom et al., 2000) in modeling studies. However, the process-based ecosystem models always involve a great amount of parameters which needs to be considered in sensitivity analysis (Haefner, 2005; Tang and Zhuang, 2009). So it usually involves numerous simulations for global sensitivity and uncertainty analysis (Tang and Zhuang, 2009). This method usually is feasible for site-level studies, yet is impracticable for regional simulation because of the large amount of parameters and a great amount of time involved in each individual regional simulation. Local sensitivity and uncertainty analyses provide an alternative for regional scale estimation of uncertainties involved in ecosystem-level studies. The local sensitivity analysis is widely used to analyze the behavior of system in response to local parameters (Cacuci, 2003); the local uncertainty analysis is used to evaluate the uncertainties caused by specific processes or parameters (Cacuci, 2003).

A combination of local sensitivity analysis and local uncertainty analysis might be a better option for regional estimation of uncertainties involved in regional CH_4 and N_2O fluxes. Combined with the Bayesian-based Markov Chain Monte Carlo method, it might provide

reasonable evaluation of the uncertainties in simulation of terrestrial CH_4 and N_2O fluxes. Based on our previous studies which report the terrestrial CH_4 and N_2O fluxes over North America during 1979-2008 (Tian et al., 2010b; Xu et al., 2010), we proposed to develop an approach to evaluate the uncertainties involved in the regional estimations of CH_4 and N_2O fluxes over North America.

Specifically, the major objectives of this study are: 1) to propose a new approach for evaluating the parameter-induced uncertainties in modeling studies; 2) to conduct a local sensitivity analysis to identify the major parameters in the DLEM model that controls regional CH_4 and N_2O fluxes over North America; and 3) to carry out a local uncertainty analysis to estimate the uncertainties in the estimated regional CH_4 and N_2O fluxes over North America. The simulations for sensitivity and uncertainty analyses were conducted at continental scale, and the analyses were carried out at biome-, country-, and continental-levels.

2. Materials and methods

2.1. Brief description of the DLEM

The model used in this study is an integrative ecosystem model DLEM which couples major biogeochemical cycles, hydrological cycles, and vegetation dynamics to make spatially-explicit estimations of carbon, nitrogen, and water fluxes and pool sizes in terrestrial ecosystems (Lu, 2009; Ren et al., 2007a, 2007b; Tian et al., 2005, 2008, 2010a, 2010b; Zhang, 2008; Zhang et al., 2007; Xu, 2010). 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 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, US Long-Term Ecological Research 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, 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; Lu, 2009; Ren et al., 2007a, 2007b; Tian et al., 2005, 2008, 2010a, 2010b; Zhang, 2008; Zhang et al., 2007; Xu et al., 2010), and the CH_4 and N_2O modules have been described in detail in *Tian et al* (Tian et al., 2010b).

The methane module in the DLEM model mainly simulates the production, consumption, and transport of CH_4. 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, soil organic matter decomposition, and litter-fall decomposition (Tian et al., 2005). 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., 2005).

In the DLEM, both denitrification and nitrification processes are simulated as one-step process as DLEM does not consider the mid-products in each process. Nitrification, a process

converting ammonium into nitrate, is simulated as a function of soil temperature, moisture, and the NH_4^+ concentration (Lin et al., 2000; Tian et al., 2010b). Denitrification, through which the nitrate is converted into N gases, is simulated in the DLEM as a function of soil temperature, moisture, and the NO_3^- concentration (Lin et al., 2000; Tian et al., 2010b). All the products of nitrification and denitrification that leave system are N-containing gases. The empirical equation reported by Davidson et al (Davidson et al., 2000) is used to separate N_2O from other gases (mainly NO and N_2).

2.2. Parameters for analysis

In the DLEM model, there are eight parameters directly control CH_4 production, consumption, and transport, and four parameters directly control N_2O flux. The eight parameters directly controlling CH_4 flux are V_{CH4Pro} (the maximum rate of CH_4 production), K_{CH4Pro} (half-saturation coefficient for CH_4 production), $V_{CH4OxiAir}$ (the maximum rate of atmospheric CH_4 oxidation), $K_{CH4OxiAir}$ (half-saturation coefficient for atmospheric CH_4 oxidation), $V_{CH4OxiSoil}$ (the maximum rate of soil CH_4 oxidation), $K_{CH4OxiSoil}$ (half-saturation coefficient of soil CH_4 oxidation), $V_{CH4Plant}$ (the maximum rate of CH_4 oxidation during plant transport), and $K_{CH4Plant}$ (half-saturation coefficient of CH_4 oxidation during plant transport). The four parameters directly controlling N_2O flux are V_{nit} (the maximum rate of nitrification), K_{nit} (half-saturation of nitrification), V_{denit} (the maximum rate of denitrification), and K_{denit} (half-saturation of denitrification). As the local sensitivity analysis and uncertainty analysis were conducted in this study, the above eight parameters directly responsible for CH_4 flux were selected for sensitivity and uncertainty analyses for terrestrial CH_4 flux, and four parameters directly responsible for N_2O flux were selected for the sensitivity and uncertainty analyses for terrestrial N_2O flux.

2.3. Methods

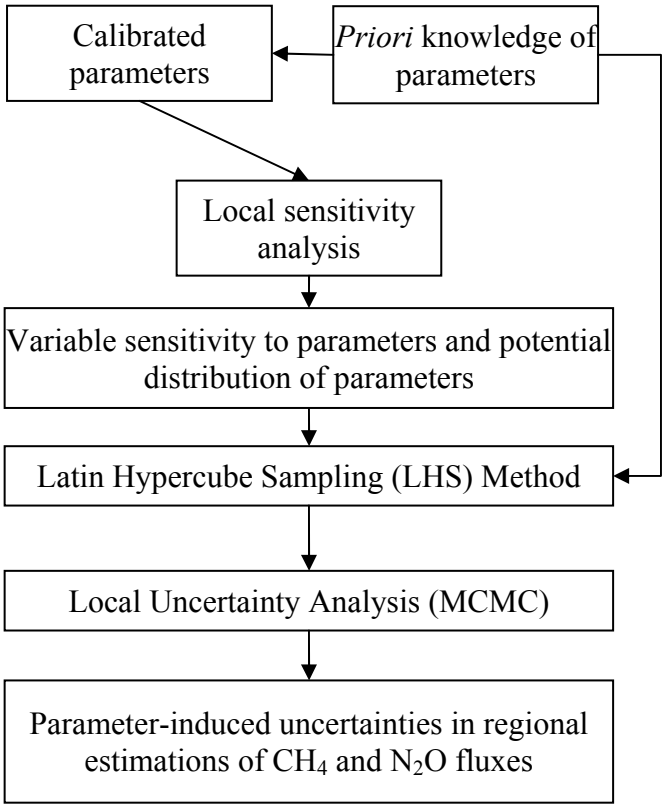


Fig. 28 Diagram showing the approach combining local sensitivity and uncertainty analyses in evaluating parameter-induced uncertainties in regional estimation of terrestrial fluxes of CH_4 and N_2O (MCMC: Markov Chain Monte Carlo)

A combination of local sensitivity analysis and local uncertainty analysis was proposed to evaluate regional terrestrial fluxes of CH_4 and N_2O, respectively (Fig. 28). First, we assumed that the distribution of each parameter follows normal distribution based on our previous calibration experience (Tian et al., 2010b). Second, we conducted local sensitivity analysis to identify the major parameters responsible for terrestrial CH_4 and N_2O fluxes, respectively, and confirm the distribution of each parameter in controlling these fluxes; the local sensitivity analysis was conducted by evaluating the changes in simulated gas fluxes in response to a 20% increase and decrease of each parameter. Third, combined with the *priori* knowledge of parameters for CH_4

and N_2O modules, we used improved Latin Hypercube Sampling (LHS) approach to randomly select an ensemble of 200 sets of 8 parameters responsible for CH_4 flux, and an ensemble of 100 sets of 4 parameters responsible for N_2O flux, in the Dynamic Land Ecosystem Model (DLEM). Finally, we set up the simulations by using the sampled 300 pairs of parameters; and the simulated results were analyzed to derive the potential uncertainties in gas fluxes induced by parameterization processes.

The LHS was selected in this study because it is a reliable approach to conduct random sampling with greatly-reduced calculation (Haefner, 2005). The algorithm is as follows: divide the range of possibility of one parameter being selected (i.e. 0 ~ 1) into 100 (or 200), equally probable regions, and then randomly (uniformly) select one value from each region of this parameter. Then the Latin Hypercube mechanism was used to pair all the sampled parameters to form an ensemble of parameters.

Finally, we used the inverse distribution function for normal distribution in SPSS software package for windows XP to transform the P-value back to their original values based on the mean and standard deviation for each parameter, respectively. The mean (μ) and standard deviation (σ) for each parameter were obtained based on the following methods. We assumed that our identified optimal value for each parameter is the mean (Tian et al., 2010b) (Eq 26), and the distance from mean to its closer boundary set by *priori* knowledge is 95% confidence interval. So the standard deviation could be inverted based on the known range and mean based on Eq 27. For example, if the priori range of K_{nit}, the half-saturation coefficient for nitrification in cropland is (1, 10), and the identified optimal parameter is 5. So the closer distance from mean to boundary is 4 (4 = 5-1), and the standard deviation is 4 / 1.96 = 2.04.

$$\boldsymbol{\mu = \beta_i} \qquad \text{Equation 26}$$

$$\sigma = \frac{\min(|\mu - B_{min}|, |B_{max} - \mu|)}{1.96} \qquad \text{Equation 27}$$

where β_i is the calibrated value for parameter i; β_{min} is the low end, and β_{max} is the high end of the priori range of parameter i, *min()* returns the minimum of the two numbers in the brackets; 1.96 is the z-value of the normal distribution at 95% significance level. Because the entire region of each parameter has been equally probable separated and selected, the randomly sampled values could completely represent the distribution of parameter population.

For each of the 17 types of biomes over North America, we set the calculated parameters based on the previously-described method (Tian et al., 2010b). For each simulation, one pair of P-value for all parameters was used for all biomes. Totally, an ensemble of 200 simulations for CH_4 flux, and an ensemble of 100 simulations for N_2O flux were set up for the time period of 1979-2008. Finally, the results were analyzed to acquire the distribution of simulated terrestrial CH_4 and N_2O fluxes over North America caused by parameter uncertainty. The 95% confidence interval for two fluxes were calculated and reported.

2.4. Statistic methods

All the statistic analyses were conducted by using SPSS and R-program (www.r-project.org) for windows XP. The improved LHS method was conducted in R-program. The *Kolmogorov-Smirnov* test was used for testing the normality of simulated fluxes of CH_4 and N_2O at continental, country-, and biome-levels.

3. Results

3.1. Sensitivity analysis of terrestrial CH_4 flux

Table 19 Sensitivity response of CH_4 to percentage changes in major parameters in DLEM CH_4 module at continental and country levels (The changes in CH_4 flux were defined as the relative change comparing to simulated results with unchanged parameters)

Parameters	Changes	Changes in CH_4 flux (%)	Changes in CH_4 flux (%)	Changes in CH_4 flux (%)	Changes in CH_4 flux (%)
		USA	Canada	Mexico	NA
V_{CH4Pro}	-20%	-21.13%	-22.55%	9.30%	-22.19%
	+20%	20.08%	22.21%	-6.84%	21.48%
K_{CH4Pro}	-20%	1.07%	0.68%	-2.66%	0.90%
	+20%	-1.05%	-0.65%	2.60%	-0.88%
$V_{CH4OxiAir}$	-20%	4.87%	2.80%	-99.35%	4.88%
	+20%	-4.93%	-2.80%	99.55%	-4.91%
$K_{CH4OxiAir}$	-20%	-5.02%	-2.86%	101.10%	-5.00%
	+20%	3.54%	2.03%	-71.98%	3.54%
$V_{CH4OxiSoil}$	-20%	-0.04%	0.00%	-0.11%	-0.02%
	+20%	-0.03%	0.00%	0.21%	-0.02%
$K_{CH4OxiSoil}$	-20%	-0.03%	0.00%	-0.10%	-0.01%
	+20%	-0.04%	0.00%	0.00%	-0.02%
$V_{CH4Plant}$	-20%	1.05%	0.88%	-6.65%	1.05%
	+20%	-1.09%	-0.88%	6.72%	-1.06%
$K_{CH4Plant}$	-20%	-1.20%	-1.03%	6.31%	-1.19%
	+20%	0.79%	0.70%	-4.48%	0.80%

Table 20 Sensitivity response of CH_4 to percentage changes in major parameters in DLEM CH_4 module across biomes (The changes in CH_4 flux were defined as the relative change comparing to simulated results with unchanged parameters)

Parameters	Changes	Changes in CH_4 flux (%)								
		Tundra	Boreal forest	Temperate forest	Tropical forest	Shrub	Grassland	Desert and others	Crop	Wetland
V_{CH4Pro}	-20%	3.19%	5.73%	4.30%	2.20%	1.18%	1.43%	5.06%	3.19%	-17.84%
	+20%	-3.17%	-5.72%	-4.30%	-2.20%	-1.19%	-1.43%	-5.06%	-3.18%	17.29%
K_{CH4Pro}	-20%	-0.25%	-1.52%	-1.05%	-0.57%	-0.34%	-0.09%	-1.16%	-0.58%	0.66%
	+20%	0.24%	1.37%	0.95%	0.51%	0.30%	0.08%	1.05%	0.52%	-0.62%
$V_{CH4OxiAir}$	-20%	-23.17%	-25.73%	-24.30%	-22.19%	-21.19%	-21.39%	-25.06%	-23.19%	0.10%
	+20%	23.19%	25.73%	24.30%	22.19%	21.18%	21.39%	25.06%	23.18%	-0.09%
$K_{CH4OxiAir}$	-20%	23.65%	26.25%	24.79%	22.64%	21.61%	21.82%	25.57%	23.76%	-0.08%
	+20%	-16.79%	-18.64%	-17.61%	-16.08%	-15.35%	-15.51%	-18.16%	-16.85%	0.08%
$V_{CH4OxiSoil}$	-20%	0.00%	0.00%	0.00%	0.00%	-0.01%	-0.04%	0.00%	0.00%	0.00%
	+20%	0.01%	0.00%	0.00%	0.00%	0.00%	0.01%	0.00%	0.00%	0.00%
$K_{CH4OxiSoil}$	-20%	0.00%	0.00%	0.00%	0.00%	0.00%	0.01%	0.00%	0.00%	0.01%
	+20%	0.01%	0.00%	0.00%	0.00%	0.00%	-0.03%	0.00%	0.00%	0.00%
$V_{CH4Plant}$	-20%	-0.01%	-0.03%	-0.04%	-0.08%	-0.01%	-0.03%	-0.05%	-0.04%	0.88%
	+20%	0.02%	0.03%	0.04%	0.08%	0.01%	0.02%	0.05%	0.03%	-0.86%
$K_{CH4Plant}$	-20%	0.02%	0.03%	0.05%	0.09%	0.01%	0.02%	0.06%	0.04%	-0.96%
	+20%	0.00%	-0.02%	-0.03%	-0.06%	-0.01%	-0.01%	-0.04%	-0.03%	0.67%

Based on the simulation experiments of local sensitivity analysis, we analyzed the response sensitivity of terrestrial CH_4 flux at continental-, country-, and biome-levels to the major parameters in CH_4 module, which were summarized in Table 19. The changes in parameters altered the country-level and continental CH_4 fluxes in different magnitudes. For example, a 20% decrease in V_{CH4Pro}, the maximum production rate of CH_4, decreased the CH_4 emission by 21.13% over the USA, 22.55% over Canada, and 22.19% over the entire North America, while increased CH_4 uptake by 9.30% over the Mexico; comparably, a 20% decrease in K_{CH4Pro}, the half-saturation coefficient CH_4 production, yielded smaller yet contrasting effects on country-level and continental level CH_4 fluxes. A 20% decrease in $V_{CH4OxiAir}$ increased CH_4 emission by 4.87% over USA, 2.80% over Canada, and 4.88% over the continental North America, while significantly increased CH_4 uptake by 99.35% over Mexico. Any changes in $V_{CH4OxiSoil}$ and $K_{CH4OxiSoil}$ did not result in substantial responses in country-level and continental CH_4 fluxes. A 20% decrease in $V_{CH4Plant}$ caused increase in CH_4 emission by 1.05% over the USA, 0.88% over Canada, and 1.05% over the continental North America, while increased CH_4 uptake by 6.65% over Mexico (Table 19). For all eight parameters, the increases and decreases in each parameters caused similar magnitude, while in contrasting directions, in the terrestrial CH_4 fluxes.

We further analyzed the response of biome-level fluxes of CH_4 flux to changes in eight parameters for nine aggregated biome types that were summarized in Table 20. The responses of CH_4 flux to changes in eight parameters varied among biomes; for example, the dominant parameters controlling CH_4 flux are $V_{CH4OxiAir}$ and $K_{CH4OxiAir}$ for tundra, boreal forest, temperate forest, tropical forest, shrub, grassland, desert, and crop, while V_{CH4Pro} for wetland. A change in two pairs of parameters, $V_{CH4OxiSoil}$ and $K_{CH4OxiSoil}$, $V_{CH4Plant}$ and $K_{CH4Plant}$, did not cause

considerable change in biome-level CH_4 fluxes for all individual biomes. Overall, the CH_4 flux from wetland responded in the most sensitive ways to the CH_4 production-related parameters, while the CH_4 fluxes from other biomes responded in the most sensitive ways to the parameters directly controlling atmospheric CH_4 oxidation (Table 20).

3.2. Sensitivity analysis of terrestrial N_2O flux

Table 21 Sensitivity response of N_2O to percentage changes in major parameters in DLEM N_2O module at continental and country levels (The changes in N_2O flux were defined as the relative change comparing to simulated results with unchanged parameters)

Parameters	Changes	Changes in N_2O flux (%)			
		USA	Canada	Mexico	NA
V_{nit}	-20%	-6.39%	-11.27%	-4.15%	-6.70%
	+20%	5.13%	9.93%	2.87%	5.43%
K_{nit}	-20%	2.05%	4.57%	1.05%	2.25%
	+20%	-1.85%	-3.84%	-0.95%	-1.99%
V_{denit}	-20%	-15.76%	-15.22%	-12.88%	-14.92%
	+20%	14.52%	13.91%	11.47%	13.63%
K_{denit}	-20%	8.64%	9.19%	5.37%	7.90%
	+20%	-6.94%	-7.46%	-4.40%	-6.38%

Table 22 Sensitivity response of N_2O to percentage changes in major parameters in DLEM N_2O module across biomes (The changes in N_2O flux were defined as the relative change comparing to simulated results with unchanged parameters)

Parameters	Changes	Changes in N_2O flux (%)								
		Tundra	Boreal forest	Temperate forest	Tropical forest	Shrub	Grassland	Desert and others	Crop	Wetland
V_{nit}	-20%	-11.94%	-12.65%	-6.20%	-6.23%	-5.91	-9.51%	-6.18%	-2.91%	-9.94%
	+20%	10.66%	11.73%	4.92%	4.50%	4.72	7.67%	4.86%	1.93%	8.37%
K_{nit}	-20%	6.04%	7.17%	1.44%	0.80%	1.96	1.41%	1.91%	2.30%	1.18%
	+20%	-4.77%	-5.89%	-1.33%	-0.77%	-1.61	-1.32%	-1.78%	-2.17%	-1.06%
V_{denit}	-20%	-15.95%	-10.34%	-18.30%	-18.76%	-7.78	-11.65%	-17.64%	-17.56%	-13.49%
	+20%	14.91%	8.12%	17.39%	18.05%	5.98	9.49%	16.66%	16.66%	12.41%
K_{denit}	-20%	11.31%	7.54%	5.64%	3.65%	4.38	6.39%	7.27%	12.38%	6.59%
	+20%	-8.68%	-6.42%	-4.80%	-3.24%	-3.57	-5.36%	-5.95%	-9.63%	-5.26%

Table 21 summarizes the response sensitivities in N_2O flux to changes in four parameters in N_2O module. The changes in parameters altered the country-level and continental N_2O fluxes, in different magnitude. For example, a 20% decrease in V_{nit}, the maximum rate of nitrification, decreased the N_2O emission by 6.39% over the USA, 11.27% over Canada, 4.15% over Mexico, and 6.70% over the entire North America; a 20% increase in V_{denit}, the maximum rate of denitrification, increase N_2O emission by 14.52% over the USA, 13.91% over the Canada, 11.47% over Mexico, and 13.63% over the entire continental North America (Table 21). For all four parameters, the increases and decreases in each parameters caused similar magnitude, while in contrasting directions, in the terrestrial N_2O fluxes.

The response of biome-level N_2O fluxes to changes in parameters for nine aggregated biome types were summarized in Table 22. The response sensitivity of N_2O flux to changes in four parameters is consistent across biomes; for example, the major parameter controlling N_2O flux is V_{denit}, followed by K_{denit}; meanwhile, the parameters for nitrification also played an important role in influencing N_2O fluxes in nine aggregated biome types (Table 22).

3.3. Estimated uncertainties in terrestrial CH_4 flux

Table 23 Means and standard deviation of major parameters for CH_4 in the DLEM (standard deviations in brackets)

Major ecosystem type	$V_{CH4ProMax}$ (g C m^{-3} day^{-1})	$V_{CH4OxidairMax}$ (g C m^{-3} day^{-1})	$V_{CH4Oxidtrans}$ (g C m^{-3} day^{-1})	$V_{CH4Oxidsoilmax}$ (g C m^{-3} day^{-1})	$Km_{CH4Prod}$ (g C m^{-3})	$Km_{CH4Oxidair}$ (ppm)	$Km_{CH4Oxidtrans}$ (g C m^{-3})	$Km_{CH4Oxidsoil}$ (g C m^{-3})
Tundra	0.25 (0.13)	0.085 (0.04)	0.1 (0.05)	0.1 (0.05)	10 (4.24)	10 (2.55)	2.5 (1.11)	3 (1.36)
Boreal broad leaf deciduous forest	0.35 (0.18)	0.08 (0.04)	0.1 (0.05)	0.1 (0.05)	10 (4.24)	10 (2.55)	2.5 (1.11)	3 (1.36)
Boreal needle leaf evergreen forest	0.35 (0.18)	0.071 (0.04)	0.1 (0.05)	0.1 (0.05)	10 (4.24)	10 (2.55)	2.5 (1.11)	3 (1.36)
Temperate broad leaf deciduous forest	0.25 (0.13)	0.042 (0.02)	0.2 (0.1)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Temperate broad leaf evergreen forest	0.4 (0.20)	0.027 (0.014)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Temperate needle leaf evergreen forest	0.65 (0.33)	0.039 (0.020)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Tropical dry forest	0.5 (0.25)	0.02 (0.01)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Tropical rain forest	0.45 (0.23)	0.015 (0.008)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Temperate mixed forest	0.65 (0.33)	0.048 (0.024)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Deciduous shrub	0.5 (0.25)	0.031 (0.016)	0.25 (0.13)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Evergreen shrub	0.25 (0.13)	0.02 (0.01)	0.2 (0.10)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
C3 grassland	0.5 (0.25)	0.03 (0.015)	0.2 (0.10)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
C4 grassland	0.6 (0.30)	0.02 (0.01)	0.1 (0.05)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)
Herbaceous wetland	1.45 (0.72)	0.032 (0.016)	5 (2.55)	2.5 (1.28)	5 (1.69)	10 (2.55)	3.5 (1.62)	3.5 (1.62)
Woody wetland	0.55 (0.26)	0.032 (0.016)	5 (2.55)	2.5 (1.28)	5 (1.69)	10 (2.55)	3.5 (1.62)	3.5 (1.62)
Cropland (dry land)	0.4 (0.20)	0.02 (0.01)	0.3 (0.15)	0.35 (0.18)	15 (6.80)	10 (2.55)	10 (4.93)	12 (5.95)
Desert	0.25 (0.13)	0.05 (0.026)	0.25 (0.128)	0.1 (0.05)	15 (6.80)	10 (2.55)	2.5 (1.11)	3 (1.36)

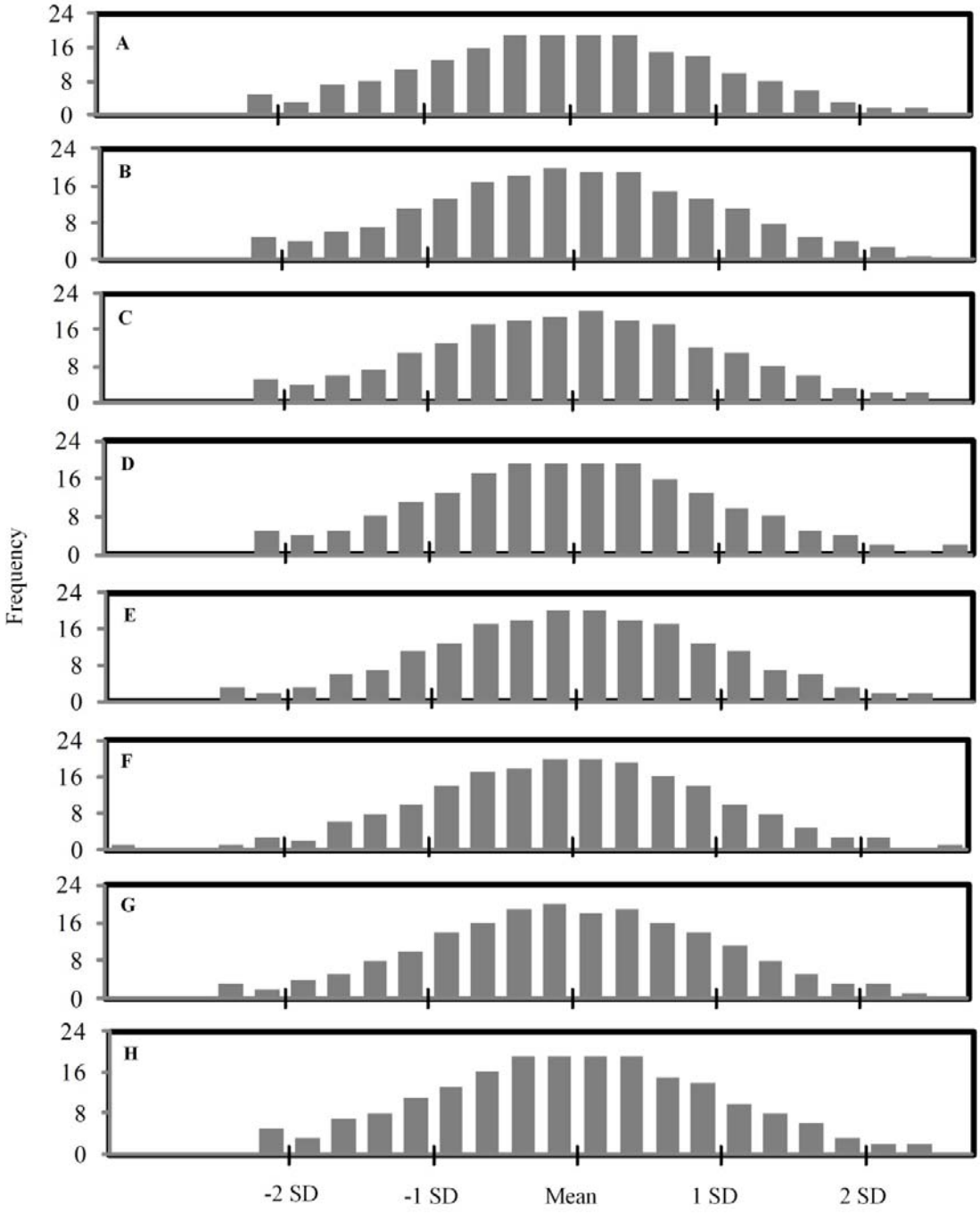


Fig. 29 Distribution of sampled major parameters for CH_4 module (A: V_{CH4Pro}; B: K_{CH4Pro}; C: $V_{CH4OxiAir}$; D: $K_{CH4OxiAir}$; E: $V_{CH4OxiSoil}$; F: $K_{CH4OxiSoil}$; G: $V_{CH4Plant}$; H: $K_{CH4Plant}$; see the main text for detail definition of each parameter; the frequency is out of 200 samples)

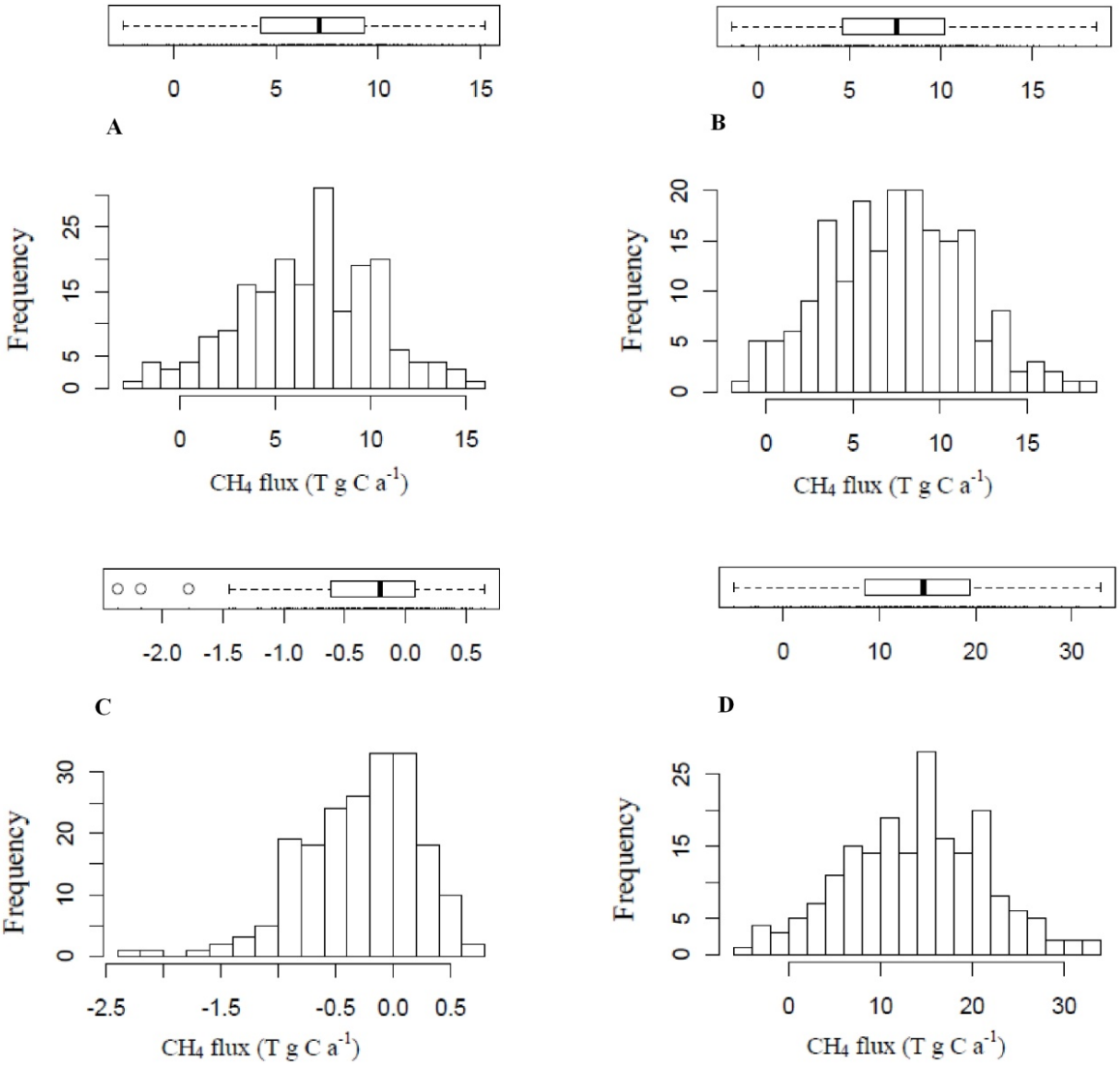


Fig. 30 Histograms and boxplots showing uncertainties of simulated CH_4 flux at both continental and country levels (A: USA; B: Canada; C: Mexico; D: North America; Downward solid triangles represent maximum values, downward open triangles represent 99% values, open circles represent means, upward open triangles represent 1% values, and upward solid triangles represent minimum values, of the simulated population of CH_4 fluxes; all the population of CH_4 fluxes for continental North America and three countries followed normal distribution according to statistical analysis)

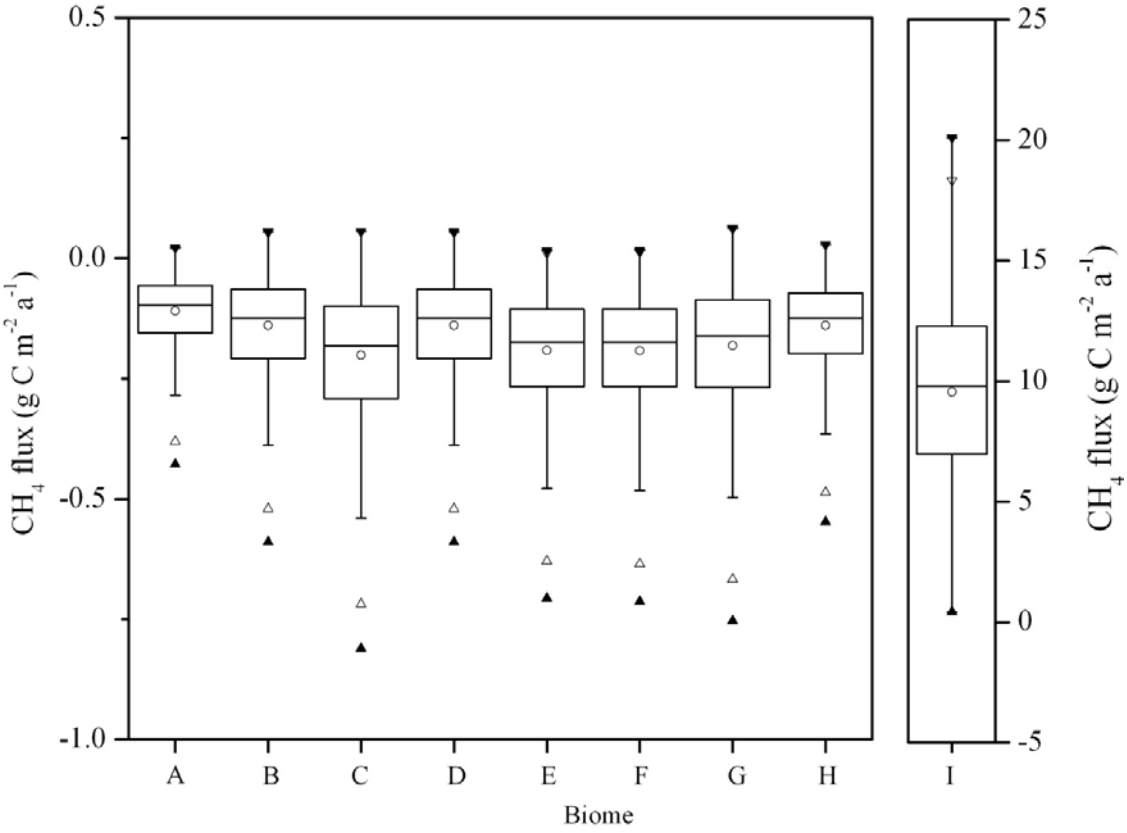


Fig. 31 Boxplots showing uncertainties of simulated CH_4 flux across biomes (A: Tundra; B: Boreal forest; C: Temperate forest; D: Tropical forest; E: Shrub; F: Grassland; G: Desert and others; H: Cropland; I: Wetland; all the population of CH_4 fluxes for nine biomes followed normal distribution according to statistical analysis)

We further conducted a local uncertainty analysis to evaluate the parameter-induced uncertainties in biome-, country-levels and continental terrestrial CH_4 fluxes. Based on the mean and standard deviation of each parameter for all biomes (Table 23), an ensemble of 200 pairs of parameters for CH_4 module was used for the uncertainty analysis. The distributions of the normalized sampled parameters are summarized in Fig. 29.

Fig. 30 shows the distribution of country- and continental levels CH_4 fluxes summarized from 200 simulations. Statistical analysis show that the simulated population of CH_4 fluxes followed normal distribution at continental scale ($P = 0.84$), and country-level for USA ($P = 0.80$), Canada ($P = 0.99$), and Mexico ($P = 0.32$). The 30-year averaged CH_4 flux was 13.89 ±

0.55 T g C a^{-1} (Mean ± SE) over the entire North America, which was not significantly different from our previously simulated result at 14.69 ± 1.64 T g C a^{-1} (Tian et al., 2010b). The 30-year averaged CH_4 flux derived from the 200 simulations was 6.71 ± 0.26 T g C a^{-1} over the USA, which is slightly lower than our previously simulated result at 7.16 ± 0.58 T g C a^{-1} (Tian et al., 2010b), was 7.46 ± 0.28 T g C a^{-1} over the Canada, which is not significantly different from our previously simulated result at 7.68 ± 1.59 T g C a^{-1} (Tian et al., 2010b). The 30-year averaged CH_4 flux derived from the 200 simulations is -0.29 ± 0.04 T g C a^{-1} over the Mexico, which is significantly larger than our previously simulated result at -0.15 ± 0.03 T g C a^{-1} (Tian et al., 2010b). Overall, the estimated 95% confidence interval of terrestrial flux of CH_4 over North America, based on this study, is (5.95 T g C a^{-1}, 23.10 T g C a^{-1}).

Fig. 31 shows the biome-level CH_4 fluxes derived from the 200 simulations. Due to the changes in biome area over study period, the areal averaged flux was presented. A statistical analysis found that the simulated population of biome-level CH_4 fluxes followed normal distribution; the significances for the normality of simulated population of CH_4 flux are P = 0.33 for tundra, P = 0.39 for boreal forest, P = 0.19 for temperate forest, P = 0.39 for tropical forest, P = 0.28 for shrub, P = 0.28 for grassland, P = 0.34 for desert, P = 0.28 for cropland, 0.92 for wetland, respectively. It could be concluded that all the simulated population of CH_4 fluxes for each biome follows normal distribution.

3.4. Estimated uncertainties in terrestrial N_2O flux

Table 24 Means and standard deviation of major parameters for N_2O in the DLEM (standard deviations in brackets)

Major ecosystem type	$V_{denimax}$ (g $Nm^{-3}day^{-1}$)	K_{deni} (g Nm^{-3})	V_{nitmax} (g $Nm^{-3}day^{-1}$)	K_{nit} (g Nm^{-3})
Tundra	0.03 (0.015)	0.15 (0.017)	0.008 (0.004)	1 (0.403)
Boreal broad leaf deciduous forest	0.013 (0.007)	0.035 (0.076)	0.0025 (0.001)	1 (0.403)
Boreal needle leaf evergreen forest	0.05 (0.026)	0.05 (0.068)	0.003 (0.002)	1 (0.403)
Temperate broad leaf deciduous forest	0.012 (0.006)	0.15 (0.017)	0.0025 (0.001)	1 (0.403)
Temperate broad leaf evergreen forest	0.007 (0.004)	0.75 (0.289)	0.03 (0.015)	1 (0.403)
Temperate needle leaf evergreen forest	0.012 (0.006)	0.15 (0.017)	0.01 (0.005)	1 (0.403)
Tropical dry forest	0.008 (0.004)	0.25 (0.034)	0.004 (0.002)	1 (0.403)
Tropical rain forest	0.0065 (0.003)	0.15 (0.017)	0.006 (0.003)	1 (0.403)
Temperate mixed forest	0.012 (0.006)	0.15 (0.017)	0.01 (0.005)	1 (0.403)
Deciduous shrub	0.055 (0.028)	0.5 (0.16)	0.005 (0.003)	1 (0.403)
Evergreen shrub	0.16 (0.082)	0.75 (0.289)	0.0025 (0.0013)	1 (0.403)
C3 grassland	0.055 (0.028)	0.75 (0.289)	0.005 (0.003)	1 (0.403)
C4 grassland	0.035 (0.018)	0.75 (0.289)	0.0035 (0.0018)	1 (0.403)
Herbaceous wetland	0.007 (0.0036)	0.5 (0.162)	0.005 (0.0026)	1 (0.403)
Woody wetland	0.0013 (0.0007)	0.35 (0.085)	0.005 (0.0026)	1 (0.403)
Cropland (dry land)	0.052 (0.027)	4.5 (1.79)	0.25 (0.13)	5 (2.04)
Desert	0.01 (0.005)	0.05 (0.068)	0.005 (0.0026)	1 (0.403)

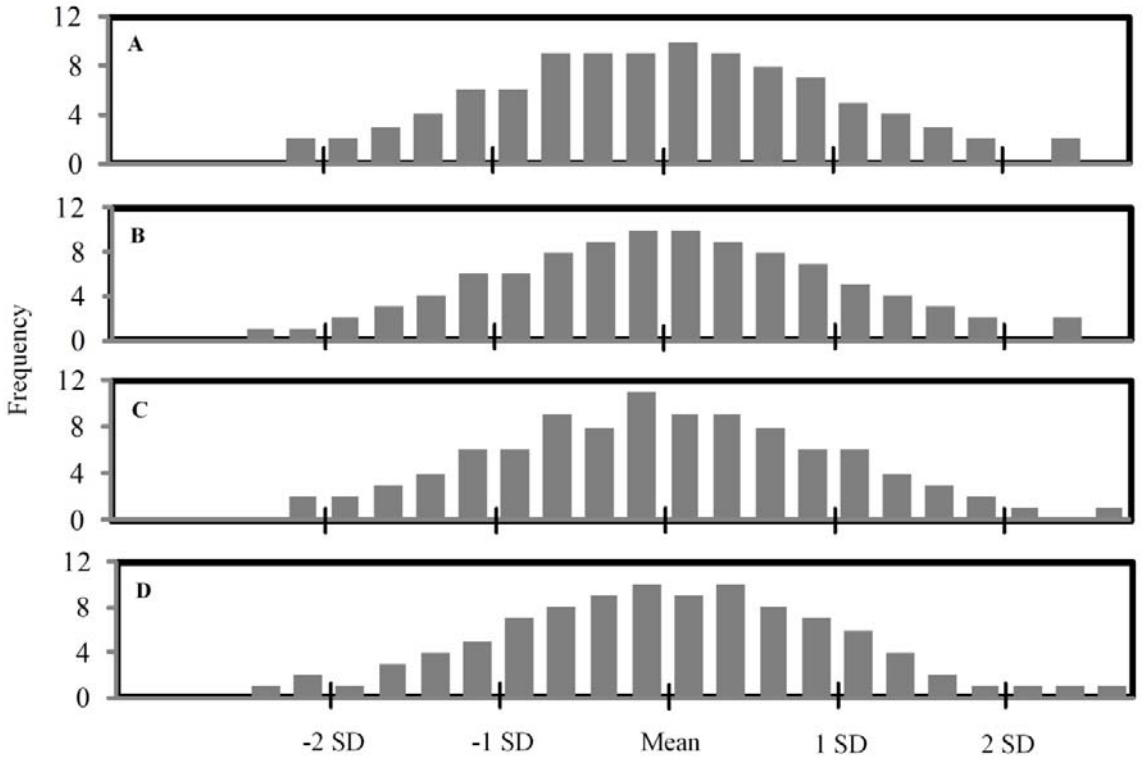


Fig. 32 Distribution of sampled major parameters for N_2O module (A: V_{nit}; B: K_{nit}; C: V_{denit}; D: K_{denit}; see the main text for detail definition of each parameter; the frequency is out of 100 samples)

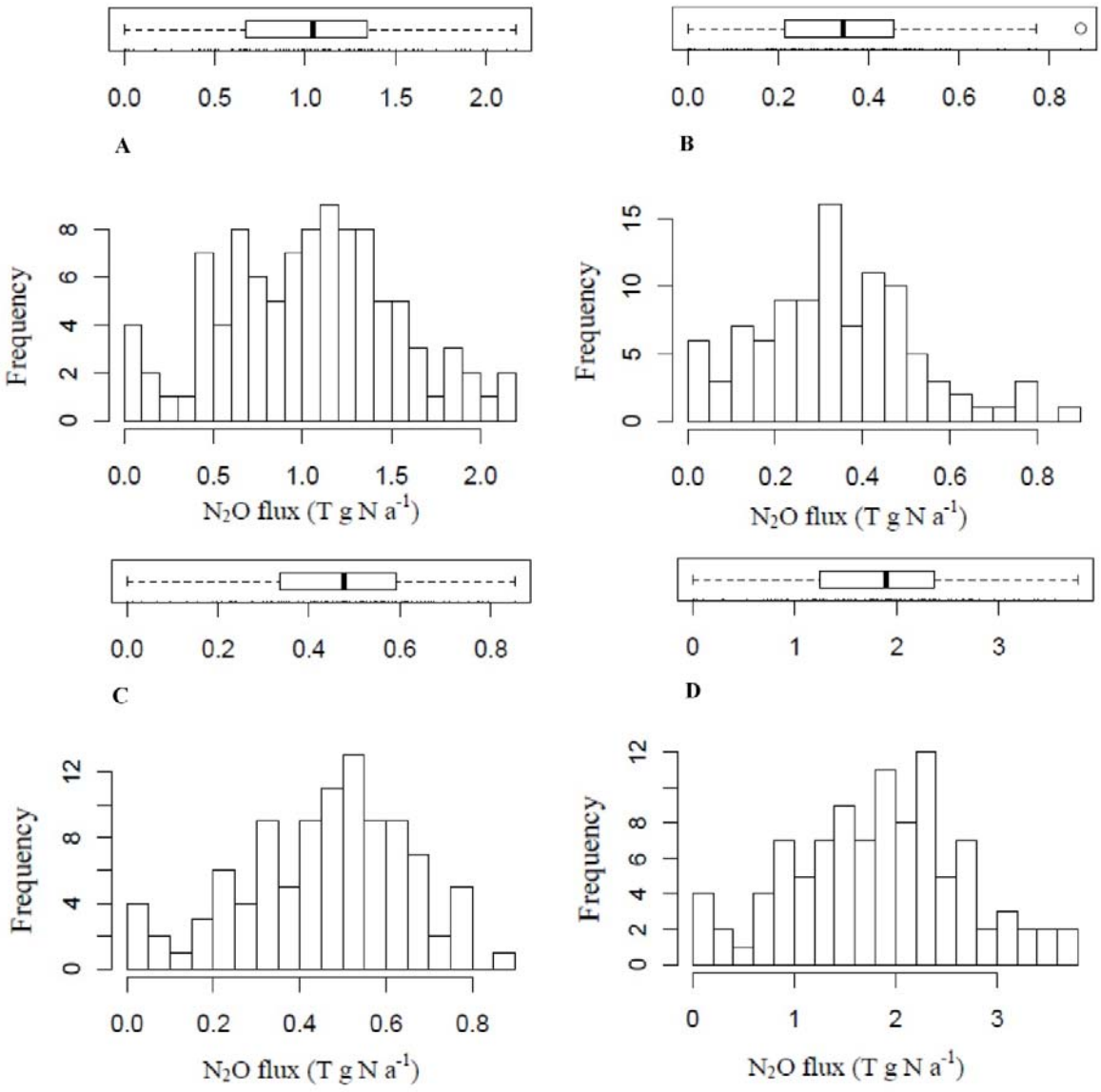


Fig. 33 Histograms and boxplots showing uncertainties of simulated N_2O flux at both continental and country levels (A: USA; B: Canada; C: Mexico; D: North America; Downward solid triangles represent maximum values, downward open triangles represent 99% values, open circles represent means, upward open triangles represent 1% values, and upward solid triangles represent minimum values, of the simulated population of N_2O fluxes; all the population of N_2O fluxes for continental North America and three countries followed normal distribution according to statistical analysis)

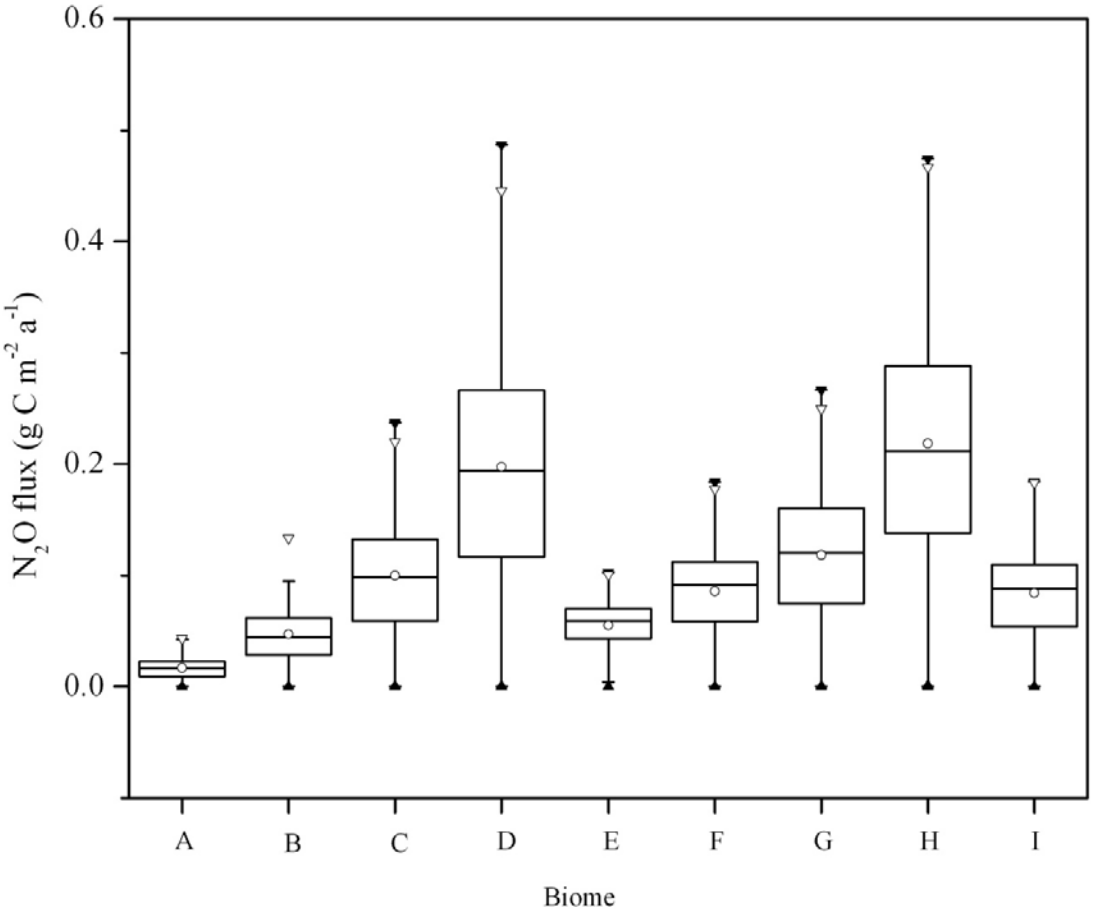


Fig. 34 Boxplots showing uncertainties of simulated N_2O flux across biomes (A: Tundra; B: Boreal forest; C: Temperate forest; D: Tropical forest; E: Shrub; F: Grassland; G: Desert and others; H: Cropland; I: Wetland; all the population of CH_4 fluxes for nine biomes followed normal distribution according to statistical analysis)

Similar to CH_4 flux, we also evaluated the uncertainties in terrestrial N_2O fluxes at biome-, country- and continental levels. Based on the mean and standard deviation of each parameter for all biomes (Table 24), an ensemble of 100 pairs of parameters for N_2O module was used for the uncertainty analysis. The distributions of the normalized sampled parameters were summarized in Fig. 32.

Fig. 33 shows the distribution of CH_4 fluxes at country- and continental levels summarized from 100 simulations. Statistical analysis show that the simulated ensemble of N_2O

fluxes followed normal distribution at continental level (P = 0.93), and country-level for the USA (P = 0.99), Canada (P = 0.80), and Mexico (P = 0.75). The 30-year averaged N_2O flux derived from the 100 simulations is 1.84 ± 0.09 T g N a^{-1} (Mean ± SE) over the entire North America, which is not significantly different from our previously simulated result at 1.94 ± 0.16 T g C a^{-1} (Tian et al., 2010b). The 30-year averaged N_2O flux derived from the 100 simulations is 1.04 ± 0.05 T g N a^{-1} over the USA, which is not significantly different from our simulated result at 1.09 ± 0.08 T g N a^{-1} (Tian et al., 2010b), 0.34 ± 0.02 T g N a^{-1} over the Canada, which is not significantly different from our previously simulated result at 0.35 ± 0.04 T g N a^{-1} (Tian et al., 2010b), and 0.46 ± 0.02 T g N a^{-1} over the Mexico, which is not significantly different from our previously simulated result at 0.50 ± 0.08 T g N a^{-1} (Tian et al., 2010b). Overall, the 95% confidence interval of 30-year averaged terrestrial N_2O flux over North America is (0.75 T g N a^{-1}, 3.38 T g N a^{-1}).

Fig. 34 shows the biome-level areal averaged N_2O fluxes derived from 100 simulations. One outlier was observed for N_2O flux from boreal forest, and there are no outliers for N_2O fluxes from all other biomes. The tropical forest, followed by cropland, is the largest source for uncertainty in N_2O flux. A statistical analysis found that the simulated population of biome-level N_2O fluxes followed normal distribution; the significances for the normality of simulated population of CH_4 flux are P = 0.66 for tundra, P = 0.50 for boreal forest, P = 0.81 for temperate forest, P = 0.95 for tropical forest, P = 0.35 for shrub, P = 0.48 for grassland, P = 0.96 for desert, 0.98 for cropland, 0.96 for wetland, respectively.

3.5. Temporal variations of uncertainties in terrestrial fluxes of CH_4 and N_2O over North America

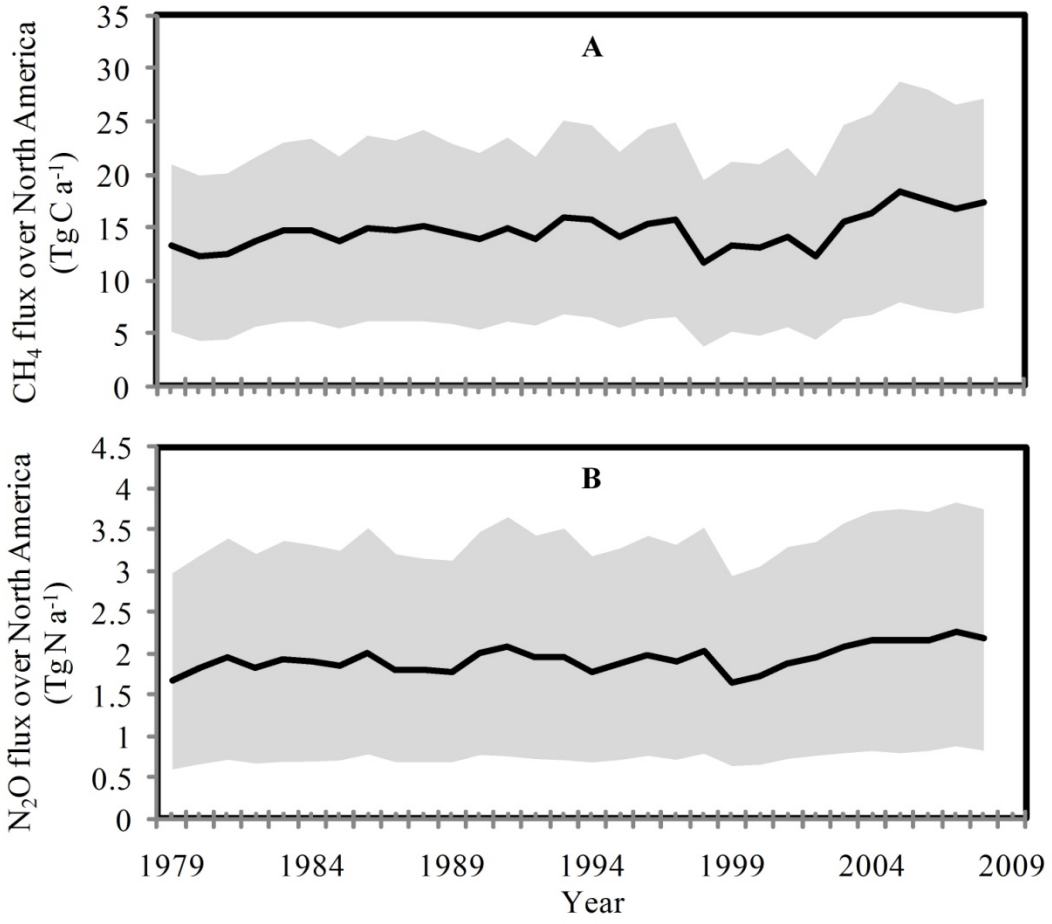


Fig. 35 Temporal variations of terrestrial fluxes of (A) CH_4 and (B) N_2O over North America and their 95% confidence boundaries during 1979-2008

After removing the outliers in simulated fluxes, we further analyzed the 95% confidence boundaries of terrestrial fluxes of CH_4 and N_2O in the past three decades. Fig. 35a and 35b show the simulated fluxes and their 95% confidence intervals in each year. Simulated results indicated that very large uncertainties existed in both CH_4 and N_2O fluxes. The simulated 95% possibility of CH_4 flux could be as low as 3.70 T g C a^{-1} or as high as 20.78 T g C a^{-1}, and the simulated 95% possibility of N_2O flux could be as low as 0.61 T g C a^{-1} or as high as 2.94 T g C a^{-1}.

Comparing the simulated fluxes and uncertainty results, the coefficient of variance is 29.78% for CH_4 flux, and 34.54% for N_2O flux. This difference indicates that the DLEM-derived CH_4 flux is more reliable than N_2O in this study.

3.6. Spatial distribution of mean and standard deviation of terrestrial CH_4 and N_2O fluxes

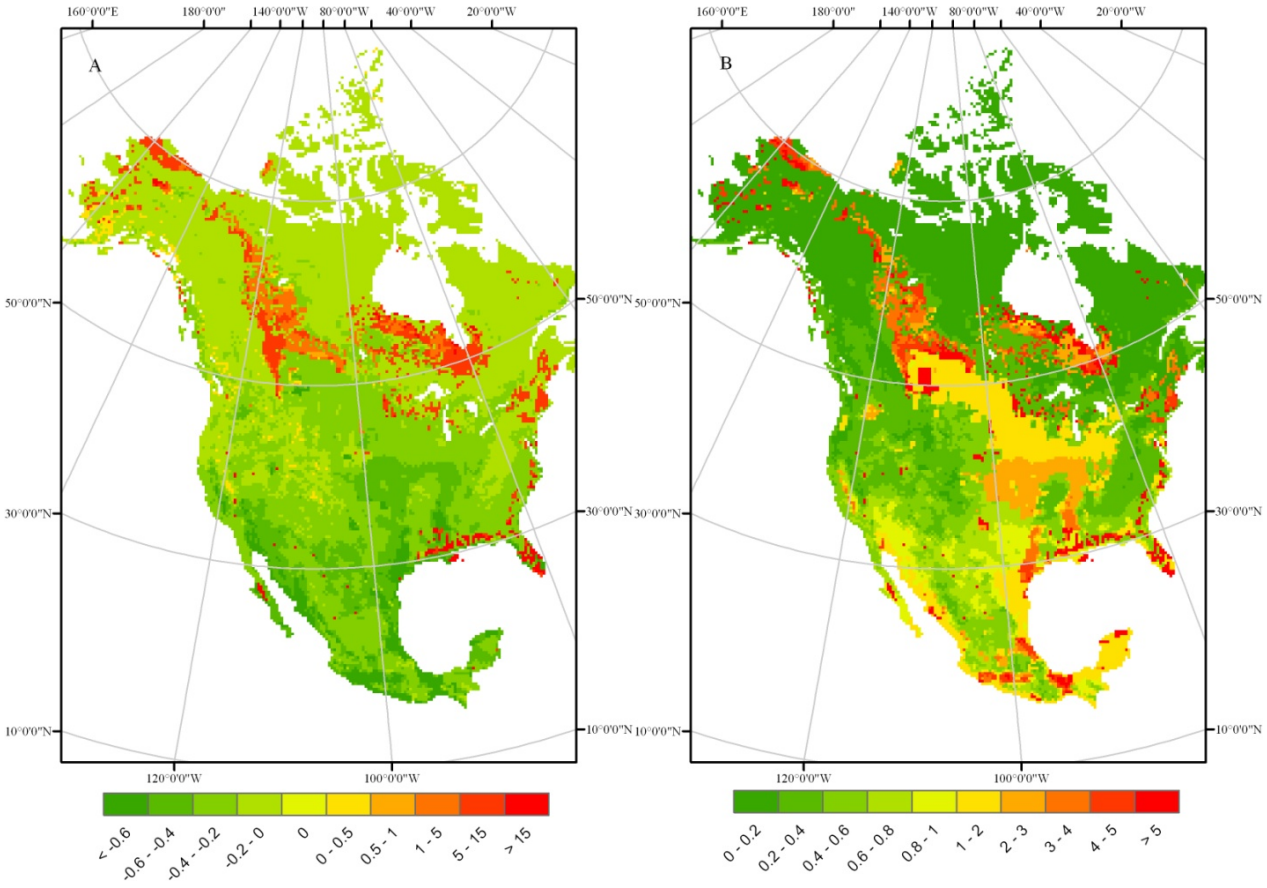


Fig. 36 Spatial distribution of mean and standard deviation of simulated annual CH_4 flux across the continental North America (A: Mean; B: Standard deviation)

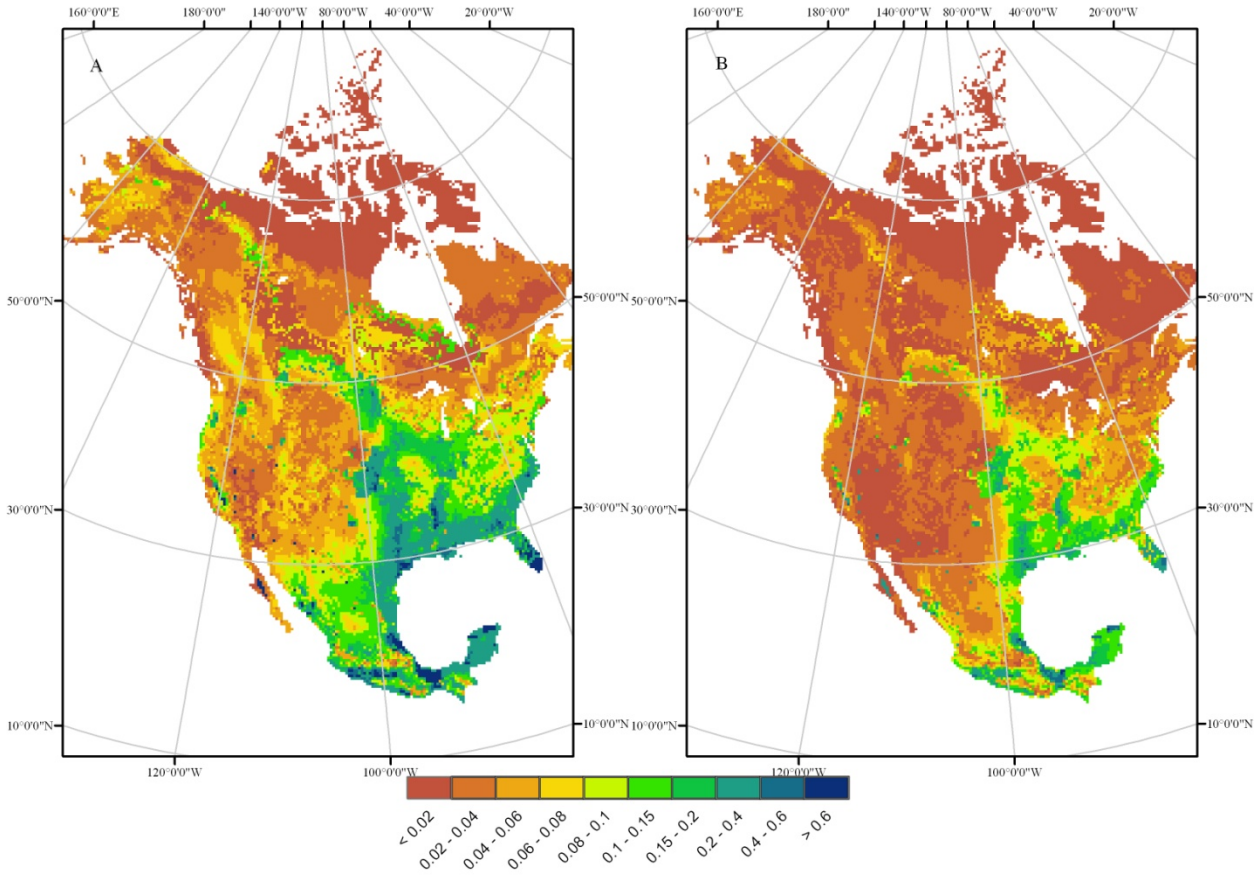


Fig. 37 Spatial distribution of mean and standard deviation of simulated annual N_2O flux across the continental North America (A: Mean; B: Standard deviation)

Based on the 200 simulation for regional CH_4 flux, and 100 simulations for regional N_2O flux, we calculated the mean and standard deviation of 30-year averaged terrestrial fluxes of CH_4 and N_2O over the continental North America. Fig. 36a and 36b show the spatial patterns of mean and standard deviation of regional CH_4 flux over the continental North America. The major sources of CH_4 locate in Alaska, Alberta in Canada, Hudson Bay Lowland, the Great Lakes area, southern Florida, and Louisiana Delta (Fig. 36a). These areas also feature the substantial uncertainties in CH_4 flux (Fig. 36b). Meanwhile, the high uncertainties in CH_4 flux also locate in part of Mexico and central conterminous US.

Fig. 37a and 37b show the mean and standard deviation of regional N_2O flux over the continental North America. The spatial distributions of uncertainties in N_2O are consistent with the mean flux of N_2O across the North America; the strong sources and high uncertainties in N_2O flux are primarily located in southeastern US, including Louisiana, Mississippi, Alabama, Georgia, Florida, North and South Carolina, Tennessee, and Arkansas; all the other areas feature relatively low uncertainties in N_2O emission (Fig. 37b).

4. Discussions

4.1. Statistic distribution of parameters for CH_4 and N_2O modules

In this study, we assumed that all the parameters for CH_4 and N_2O modules follow normal distribution. This is different from one previous study which assumes uniform distribution of parameter (Tang and Zhuang, 2009). The reason for the assumption of normal distribution for each parameter is that the behavior of most systems in reality is not linear (Haefner, 2005; Meadows, 2008); their behavior are convergent toward one or two points; around these points/states, the system behavior is following normal distributed (Haefner, 2005). Independently, the sensitivity analysis concluded that the increases and decreases in each parameter generated the terrestrial fluxes of CH_4 and N_2O in similar magnitude while in contrasting direction (Tables 19, 20, 21 and 22), which confirmed the normal distribution assumption in this study. Furthermore, the uncertainty analysis generated the normally-distributed fluxes of CH_4 and N_2O at biome-, country- and continental levels (Fig. 30, 31, 33, and 34).

4.2. Processes controlling terrestrial CH_4 flux

Sensitivity analysis is a powerful tool to examine how sensitive a model responds to parameters, input data, or other factors (Haefner, 2005). The change in model output in response to parameter alterations was defined as the model sensitivity, individually or interactively. In this study, the sensitivity analysis was used to identify the major parameters responsible for the regional CH_4 and N_2O fluxes, which could be used to reveal the major processes controlling regional gas fluxes.

The different sensitivities of biome-level CH_4 fluxes to eight parameters might be caused by the different processes controlling the CH_4 flux in specific ecosystem. For example, the CH_4 production is usually higher than CH_4 oxidation in wetlands (Conrad, 1996; Conrad and Klose, 1999), so the CH_4 production usually controls the CH_4 flux over wetland. Thus, the CH_4 flux from wetland shows the most sensitive response to CH_4 production-related parameters (Table 19). While the atmospheric CH_4 oxidation dominates the CH_4 flux over forest, desert, and grassland (Bender and Conrad, 1992; Liu et al., 2007; Saari et al., 2004; Strieg et al., 1992), so the CH_4 flux over these ecosystems exerts high sensitivity to CH_4 oxidation-related parameters, such as the $V_{CH4OxiAir}$, the maximum rate of atmospheric CH_4 oxidation. These different response sensitivities of CH_4 flux to parameters might explain the country-level differences in sensitivity analysis. For instance, the CH_4 fluxes in USA and Canada are more sensitive to V_{CH4Pro}, the maximum rate of CH_4 production, than to other parameters; this phenomenon is because the USA and Canada feature a great amount of wetlands (Mitsch and Gosselink, 2007); while the CH_4 flux in Mexico are more sensitive to CH_4 oxidation-related parameters, such as $V_{CH4OxiAir}$, the maximum rate of atmospheric CH_4 oxidation, and $K_{CH4OxiAir}$, the half-saturation coefficient for atmospheric CH_4 oxidation, than to other parameters; this might be explained by the fact that

Mexico features more dry land ecosystems, such as forest, cropland, and desert (Bridgham et al., 2006; Leopold, 1950).

4.3. Processes controlling terrestrial N_2O flux

Nitrification and denitrification are tightly coupled because the products of nitrification are the substance of denitrification (Arth et al., 1998; Conrad, 1996; Delwiche, 1981; Sheldon and Barnhart, 2009). Although several studies observed soil as sink of N_2O in limited sites (Chapuis-Lardy et al., 2007), most of the biomes function as sources, strong or weak, for N_2O at large spatial scale and over long time period (Potter et al., 1996; Williams et al., 1992). The direct production of N_2O is primarily from denitrification, so the denitrification-related parameters are major controller, while the nitrification-related parameters are minor controller on N_2O flux. For example, the denitrification predominantly controls N_2O flux over all biomes (Chapuis-Lardy et al., 2007; Conrad, 1996); thus, the V_{denit}, maximum rate of denitrification, is the major parameter yielding strong impact on N_2O flux as simulated by the DLEM model.

Overall, N_2O fluxes from all biomes are more sensitive to denitrification-related parameters than to nitrification-related parameters (Table 21), which is consistent with country-level results as shown in Table 20.

4.4. Contribution of parameter prior knowledge to uncertainties in terrestrial CH_4 and N_2O fluxes

The uncertainty analysis is the process to assess the parameter-induced uncertainties in simulated results (Cacuci, 2003). The parameter-induced uncertainties in CH_4 and N_2O fluxes varied among biomes, which might be caused by either the different dominant processes for CH_4 and N_2O fluxes, or the *priori* knowledge in determination of each parameter. The inter-biome variations in uncertainties in CH_4 and N_2O fluxes could be explained by the different dominant

processes; while the large uncertainties in CH_4 and N_2O fluxes within one biome imply the primary source of uncertainties in *priori* knowledge of parameters. To reduce the uncertainties in large-scale estimates of terrestrial CH_4 and N_2O fluxes, it is highly needed to reduce the uncertainties in *priori* knowledge of parameters. Given the large number of parameters for each models (Haefner, 2005), and uncertainties in each parameter, the precise and accurate estimation of parameters will be critical tasks for modeling efforts.

4.5. Improvement needs

This study proposed an approach by combining local sensitivity analysis and local uncertainty analysis to evaluate the parameter-induced uncertainties at regional scale, and then further evaluate the uncertainties in terrestrial CH_4 and N_2O fluxes over North America. This study did provide an approach for large-scale uncertainty analysis for terrestrial CH_4 and N_2O fluxes; however, several limitations of this study should be noted. First, the assumed normal distribution of parameters might not be representative of the reality; second, the uncertainty lies in the other parameters directly related to carbon or nitrogen fluxes has not been investigated, so the uncertainty might be underestimated in present study; third, the present study was built upon the assumption that the uncertainties for all biomes follow the same distribution regarding to the same set of parameters, which might need further examination.

Although local sensitivity and uncertainty analyses were conducted, the changes in local parameters might feedback to global parameters of the DLEM model (Haefner, 2005), and in turn cause alteration in simulated output in terrestrial fluxes of CH_4 and N_2O; for example, increase in CH_4 emission might cause rapid depletion of dissolved organic carbon in soil, which will in turn change the soil microbial processes (Neff and Asner, 2001). This algorithm involves a lot of processes and interaction within the model, and needs more attention in the future to

comprehensively evaluate the uncertainties involved in the modeling approach. Beside the uncertainties caused by parameters, the input data and model structure might also induce uncertainties in the regional estimation of terrestrial fluxes of CH_4 and N_2O and need further investigations.

5. Conclusions

Combined local sensitivity analysis and local uncertainty analysis, we evaluated the parameter-induced uncertainties in our regional estimates of terrestrial fluxes of CH_4 and N_2O over North America. The 95% confidence interval of terrestrial flux of CH_4 over North America, based on this study is (5.95 T g C a^{-1}, 23.10 T g C a^{-1}), and the 95% confidence interval of terrestrial flux of N_2O is (0.75 T g N a^{-1}, 3.38 T g N a^{-1}). DLEM-derived CH_4 flux is more reliable than N_2O in this study. The broad range of fluxes suggests the large uncertainties in parameters, which calls for more precise and accurate estimate of parameters in modeling efforts.

To the best of our knowledge, this study is among the first effort attempting to evaluate the uncertainty of terrestrial CH_4 and N_2O fluxes at regional scale. This study, as a preliminary attempt to find the uncertainties in regional fluxes of CH_4 and N_2O over the North America, provides a new method for uncertainty analysis for large-scale modeling efforts. The approach and the findings of different parameters in contribution to uncertainties in terrestrial CH_4 and N_2O fluxes at biome-, country- and continental levels in this study may provide information for large-scale estimation of ecosystem functioning.

Chapter 6. Conclusions

In this study, I enhanced a process-based ecosystem model by incorporating biogeochemical processes for CH_4 and N_2O production and consumption, and conducted intensive calibration for model parameterization; then I used this enhanced model to examine terrestrial fluxes of CH_4 and N_2O over terrestrial ecosystems in the continental North America during 1979-2008; I further attributed the spatial and temporal variations in terrestrial fluxes of CH_4 and N_2O to various global change factors including climate variability, O_3 pollution, nitrogen deposition, land use change, nitrogen fertilization, elevated atmospheric CO_2; finally I estimated the potential uncertainties in this study caused by parameters by using Bayesian-based Monte Carlo method.

The key conclusions are listed as below:

1) During the past 30 years, approximately 14.69 ± 1.64 T g C a^{-1} (1T g = 10^{12} g) of CH_4, and 1.94 ± 0.16 T g N a^{-1} of N_2O were released from terrestrial ecosystems in North America. At the country level, both the United States and Canada acted as CH_4 sources to the atmosphere, but Mexico mainly oxidized and consumed CH_4 from the atmosphere. Wetlands in North America contributed predominantly to the regional CH_4 source, while all other ecosystems acted as sinks for atmospheric CH_4, of which forests accounted for 36.8%. Regarding N_2O emission in North America, the United States, Canada, and Mexico contributed 56.19%, 18.23%, and 25.58%, respectively, to the continental source over the past 30 years. Forests and croplands were the two ecosystems that contributed

most to continental N_2O emission. The inter-annual variations of CH_4 and N_2O fluxes in North America were mainly attributed to year-to-year climatic variability. While only annual precipitation was found to have a significant effect on annual CH_4 flux, both mean annual temperature and annual precipitation were significantly correlated to annual N_2O flux.

2) Over the past three decades, our simulations indicates that global change factors accumulatively contributed 23.51 ± 9.61 T g CH_4-C (1Tg = 10^{12} g) emission over North America, among which ozone (O_3) pollution led to a reduced CH_4 emission by 2.30 ± 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 ± 12.42 T g CH_4-C, 0.09 ± 0.02 T g CH_4-C, 6.80 ± 0.86 T g CH_4-C, 0.01 ± 0.001 T g CH_4-C, and 3.95 ± 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 ± 7.74 T g CH_4-C.

3) 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.

4) From 1979 to 2008, the North America accumulatively emitted 58.17 ± 0.85 T g N_2O-N (1T g = 10^{12} g), of which global change factors contributed 2.81 ± 0.98 T g N_2O-N, and baseline emission contributed 55.35 ± 0.56 T g N_2O-N. The elevated CO_2 led to decreases in terrestrial N_2O emission at 0.51 ± 0.07 T g N_2O-N. Climate variability, N deposition, O_3 pollution, N fertilization, and land use conversion increased N_2O emission by 0.56 ± 0.68 T g N_2O-N, 0.50 ± 0.07 T g N_2O-N, 0.10 ± 0.02 T g N_2O-N, 0.92 ± 0.09 T g N_2O-N, and 0.16 ± 0.01 T g N_2O-N, respectively. The interactive effect among multiple factors enhanced N_2O emission by 1.10 ± 0.37 T g N_2O-N over the 30 years.
5) At country level, climate variability and elevated atmospheric CO_2 decreased, while all other single factors and multiple-factor interaction enhanced N_2O emission in the United States of America (USA) over the past 30 years; during the same time period, elevated atmospheric CO_2 and multiple factor interaction decreased, while other factors enhanced N_2O emission from Canada; and elevated atmospheric CO_2 and land conversion decreased, while other factors enhanced N_2O emission from Mexico. Southeastern part of the continental North America including central Canada, southeastern USA, and entire Mexico acted as a strong source for atmospheric N_2O while other parts functioned as a weak source of N_2O.
6) The parameter, maximum rate of CH_4 production, followed by the parameters controlling atmospheric CH_4 oxidation, is the most sensitive parameter controlling the continental CH_4 flux. The major parameters controlling biome- and country-level CH_4 fluxes varied among countries and biomes; while the ubiquitous dominant parameter controlling biome-, country-level, and continental N_2O fluxes is the maximum rate of denitrification. The 95% confidence interval of terrestrial flux of CH_4 over North America, based on this

study is (5.95 T g C a^{-1}, 23.10 T g C a^{-1}), and the 95% confidence interval of terrestrial flux of N_2O is (0.75 T g N a^{-1}, 3.38 T g N a^{-1}). The spatial distributions of the uncertainties in terrestrial CH_4 and N_2O fluxes varied across the continental North America; the largest uncertainty in CH_4 flux is from wetlands, while the largest uncertainty in N_2O flux lies in tropical forest, followed by cropland.

7) This study also found the interactive among global change factors might be very essential for environmental controls on terrestrial fluxes of CH_4 and N_2O; this calls for more field experiments to investigate the effects of multiple factor, individual and in combination, on ecosystem functioning.

Meanwhile, I identified several sources of uncertainties in current study which need to be addressed in the future study. First, for the input data, I used one set of data in this study. Various sets of input data have been developed and potentially could be used for model simulations; the evaluation of the uncertainties caused by different sources of input data will be one major method to reduce uncertainties in estimates; evaluation of the effects of various spatial scales in simulation might be another way to evaluate uncertainties. Second, for the model structure and algorithms for major processes incorporated in the DLEM model, some improvements will be helpful to reduce uncertainties; for example, explicit simulation of urban systems will greatly improve estimates; more detailed information of vertical distribution of soil profile may benefit simulation of soil biogeochemistry processes which are essential for trace gases estimations. Last, the parameterization process is one major sources of uncertainties in modeling studies; even though I have evaluated the potential uncertainties caused by parameterization process on terrestrial fluxes of CH_4 and N_2O, the present study focused on local

uncertainty analysis, global uncertainty analysis with more simulations will be necessary to narrow down the ranges of flux estimates at both continental and country-levels.

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