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Spatial and Temporal Patterns of CH4 and N2O Fluxes in Terrestrial Ecosystems of North America during 1979–2008: Application of a Global Biogeochemistry Model

Hanqin Tian, *Auburn University Main Campus*
X. Xu, *Auburn University Main Campus*
M. Liu, *Auburn University Main Campus*
Wei Ren, *Auburn University Main Campus*
C. Zhang, *Auburn University Main Campus*, et al.

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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

H. Tian[1,2], X. Xu[1,2], M. Liu[1,2], W. Ren[1,2], C. Zhang[1], G. Chen[1,2], and C. Lu[1,2]

[1]Ecosystem Dynamics and Global Ecology (EDGE) Laboratory, School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL, 36849, USA
[2]International Center for Climate and Global Change Research, Auburn University, Auburn, AL, 36849, USA

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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 $14.69 \pm 1.64\,\mathrm{T\,g\,C\,a^{-1}}$ ($1\,\mathrm{T\,g} = 10^{12}\,\mathrm{g}$) of CH_4, and $1.94 \pm 0.1\,\mathrm{T\,g\,N\,a^{-1}}$ of N_2O were released from terrestrial ecosystems in North America. At the country level, both the US 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 US, 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. 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.

Correspondence to: H. Tian
(tianhan@auburn.edu)

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 US 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., 2001a, 2001b), 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, 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, 2010; 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, 2010; 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; and (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

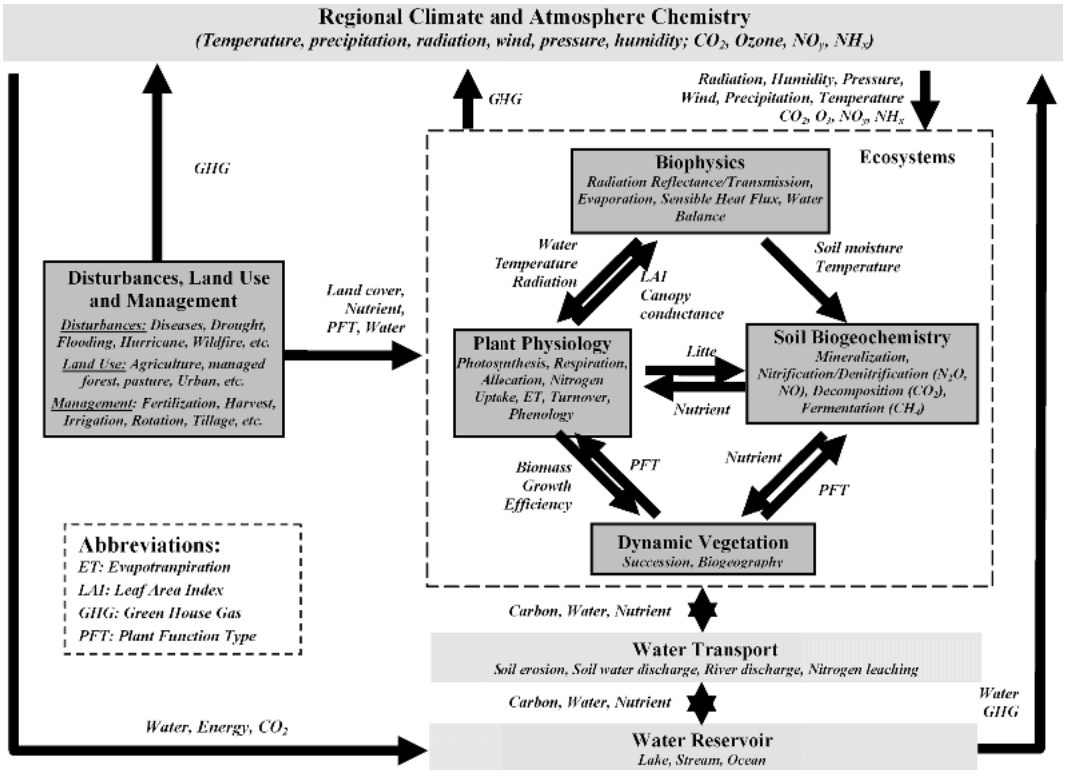

Fig. 1. Conceptual model of the Dynamic Land Ecosystem Model (DLEM) (Five core components are included in the DLEM).

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).

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, 2010; Ren et al., 2007a, 2007b, 2009; Zhang et al., 2007, 2008; Lu, 2009; Liu et al., 2008; Chen et al., 2006).

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).

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

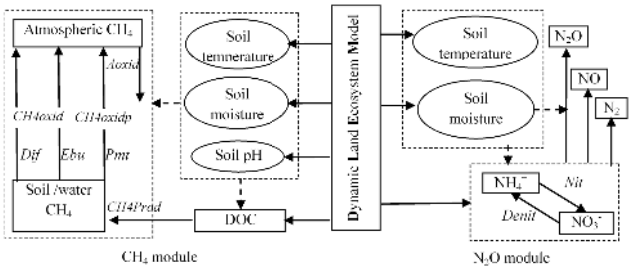

Fig. 2. Modules of CH_4 and N_2O in the Dynamic Land Ecosystem Model (DLEM) (CH_4 production, oxidation, and transport are considered in the CH_4 module; nitrification and denitrification are considered in the N_2O module).

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 leachate, 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:

$$F_{CH_4} = F_P + F_D + F_E - F_{air,\ oxid} - F_{trans,\ oxid} \quad (1)$$

where F_{CH_4} 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:

$$\frac{d[CH_4]}{dt} = f([CH_4]) \quad (2)$$
$$= CH_{4_{prod}} - \frac{F_P}{H} - \frac{F_D}{H} - \frac{F_E}{H} - CH_{4_{soil,\ oxid}}$$

where $[CH_4]$ is the concentration of CH_4 in water ($g\,C\,m^{-3}$); $CH_{4_{prod}}$ is the production of CH_4 in soil pore water ($g\,C\,m^{-3}\,d^{-1}$); $CH_{4_{soil,\ oxid}}$ 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.

CH_4 production

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

$$CH_{4_{prod}} = V_{prod,\ max} \times \frac{[DOC]}{[DOC] + Km_{prod}} \quad (3)$$
$$\times f(T_{soil}) \times f(pH) \times f_{prod}(vwc)$$

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.

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 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.

Atmospheric CH_4 oxidation

Oxidation of atmospheric CH_4 is estimated as:

$$F_{air,\ oxid} = V_{air,\ oxid,\ max} \times \frac{[Atm\ CH_4]}{[Atm\ CH_4] + Km_{air,\ oxid}} \quad (4)$$
$$\times f(T_{soil}) \times f(pH) \times f_{oxid}(vwc)$$

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 ($g\,C\,m^{-3}$);

[Atm CH_4] 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 $10\,g\,C\,m^{-2}$.

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:

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

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

Soil pore water CH_4 oxidation

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

$$CH_{4_{\text{soil, oxid}}} = \min \Big(V_{\text{soil, oxid, max}} \times \frac{[CH_4]}{[CH_4] + Km_{\text{soil, oxid}}} \times f(T_{\text{soil}}) \times f(\text{pH}) \times f_{\text{oxid}}(\text{vwc}),\ [CH_4] \Big) \quad (6)$$

where $V_{\text{soil, oxid, max}}$ and $Km_{\text{soil, oxid}}$ 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; [CH_4] is the concentration of CH_4 in soil pore water ($g\,C\,m^{-3}$).

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.

Ebullition

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

$$F_E = \max\left(([CH_4] - 6),\ 0\right) \times H \quad (7)$$

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.

Plant-mediated transport

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

$$F_P = V_{\text{plant, trans}} \times ([CH_4] - [CH_4]_{\max}) \times \min\left(\frac{\text{GPP}}{\text{GPP}_{\max}},\ 1\right) \quad (8)$$

$$[CH_4]_{\max} = [\text{Atm } CH_4] \times \beta \quad (9)$$

where F_P is the CH_4 transport via vascular plant ($g\,C\,m^{-2}\,d^{-1}$); $V_{\text{plant, trans}}$ is the transport coefficient of CH_4 transportation through plant ($m\,d^{-1}$), which is set as 0.68 (Kettunen, 2003); $[CH_4]_{\max}$ is the maximum CH_4 concentration in soil solution ($g\,C\,m^{-3}$); GPP is the gross primary productivity of current day ($g\,C\,m^{-2}\,d^{-1}$); $\text{GPP}_{\max}$ 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.

Diffusion

The DLEM treats the top 0.5 m of the soil profile as one layer, and the CH4 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.

$$F_D = V_{\text{exchange}} \times ([CH_4] - [CH_4]_{\max}) \quad (10)$$

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).

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 the Fig. 3.

In the DLEM, the effect of soil pH on methane production and oxidation (f(pH)) is calculated as a bell shape curve,

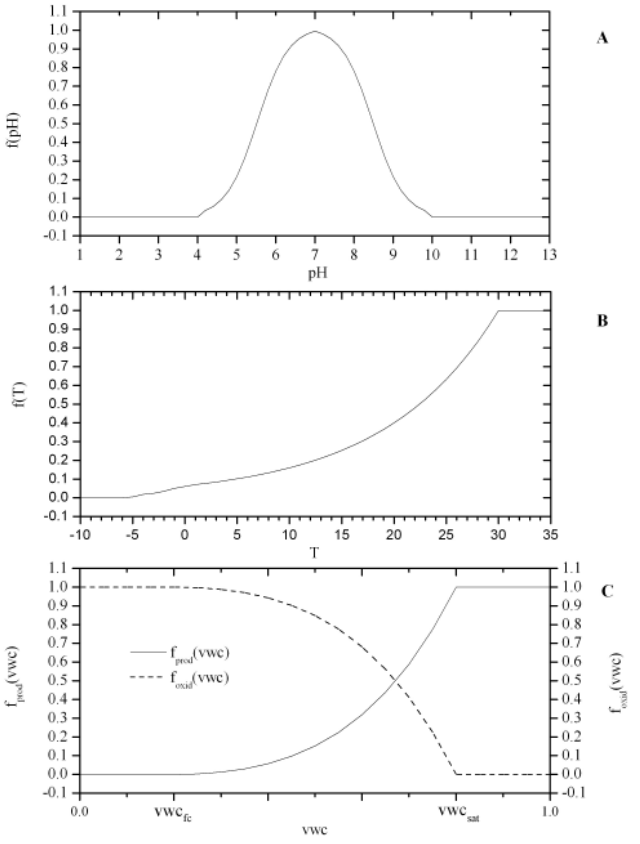

Fig. 3. Graphs showing environmental effects on methane production and oxidation – **(A)** pH effects; **(B)** Temperature effect; **(C)** Moisture effects on methane production and oxidation.

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).

$$f(\mathrm{pH}) = \begin{cases} 0 & \mathrm{pH} \le 4.0 \text{ or } \mathrm{pH} \ge 10.0 \\ \frac{1.02}{1+1\,000\,000 \times e^{(-2.5 \times \mathrm{pH})}} & 4.0 < \mathrm{pH} < 7.0 \\ \frac{1.02}{1+1\,000\,000 \times e^{(-2.5 \times (14.0-\mathrm{pH}))}} & 7.0 < \mathrm{pH} < 10.0 \end{cases} \quad (11)$$

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 (Song et al., 2009), rather than 3.

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

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.

$$f_{\mathrm{prod}}(\mathrm{vwc}) = \quad (13)$$
$$\begin{cases} 0 & \mathrm{vwc} \le \mathrm{vwc_{fc}} \\ \left(\frac{\mathrm{vwc}-\mathrm{vwc_{fc}}}{\mathrm{vwc_{sat}}-\mathrm{vwc_{fc}}}\right)^2 \times 0.368 \times e^{\left(\frac{\mathrm{vwc}-\mathrm{vwc_{fc}}}{\mathrm{vwc_{sat}}-\mathrm{vwc_{fc}}}\right)} & \mathrm{vwc_{fc}} < \mathrm{vwc} < \mathrm{vwc_{sat}} \\ 1 & \mathrm{vwc} \ge \mathrm{vwc_{sat}} \end{cases}$$

$$f_{\mathrm{oxid}}\,(\mathrm{vwc}) = 1 - f_{\mathrm{prod}}\,(\mathrm{vwc}) \quad (14)$$

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

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.

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).

$$N_{\mathrm{nit}} = \min\left(N_{\mathrm{pot,\ nit}},\ N_{\mathrm{NH_4}}\right) \quad (15)$$

$$N_{\mathrm{pot,\ nit}} = V_{\mathrm{nit,\ max}} \times \frac{N_{\mathrm{NH_4}}}{N_{\mathrm{NH_4}} + Km_{\mathrm{nit}}} \times f_{\mathrm{nit}}(T_{\mathrm{soil}}) \times f_{\mathrm{nit}}\,(\mathrm{vwc}) \quad (16)$$

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

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

where N_{nit} is the nitrification rate ($\mathrm{g\,N\,m^{-3}\,d^{-1}}$); $N_{\mathrm{pot,\ nit}}$ is the potential nitrification rate ($\mathrm{g\,N\,m^{-3}\,d^{-1}}$); $N_{\mathrm{NH_4}}$ is the concentration of NH_4^+ in the soil ($\mathrm{g\,N\,m^{-3}}$); $V_{\mathrm{nit,\ max}}$ is a parameter describing potential nitrification rate without limitation ($\mathrm{g\,N\,m^{-3}\,d^{-1}}$); Km_{nit} is the half-saturation concentration of soil NH_4^+ for the maximum nitrification rate ($\mathrm{g\,N\,m^{-3}}$); $f_{\mathrm{nit}}(T_{\mathrm{soil}})$ is a multiplier that describes the effect of soil temperature on nitrification; T_{soil} is the soil temperature (°C); $f_{\mathrm{nit}}(\mathrm{vwc})$ is a multiplier that describes the effect of water content on nitrification (Lin et al., 2000; Riedo et al., 1998); $Q_{10,\ \mathrm{nit}}$ is the temperature sensitivity of nitrification, which

is set as 2; $T_{\text{opt, nit}}$ 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.

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).

$$N_{\text{denit}} = \min\left(N_{\text{pot, denit}},\ N_{\text{NO}_3}\right) \tag{19}$$

$$N_{\text{pot, denit}} = V_{\text{denit, max}} \times \frac{N_{\text{NO}_3}}{N_{\text{NO}_3} + Km_{\text{denit}}} \times f_{\text{denit}}(T_{\text{soil}}) \times f_{\text{denit}}(\text{vwc}) \tag{20}$$

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

$$f_{\text{denit}}(\text{vwc}) = \begin{cases} 0.0 & \text{vwc} < \text{vwc}_{\text{fc}} \\ \frac{\text{vwc}}{\text{vwc}_{\text{fc}}} & \text{vwc} \geq \text{vwc}_{\text{fc}} \end{cases} \tag{22}$$

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

N_2O emission

All the products of nitrification and denitrification 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).

$$F_{\text{NO}_2} = (0.001 \times N_{\text{nitrif}} + N_{\text{denitrif}}) \times \frac{10^{\text{vwc}/Ø \times 0.026 - 1.66}}{\left(1 + 10^{\text{vwc}/Ø \times 0.026 - 1.66}\right)} \tag{23}$$

where $F_{\text{N}_2\text{O}}$ is the fluxes of N_2O from soil to the atmosphere ($\text{g N m}^{-3}\,\text{d}^{-1}$), 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 ($\text{g N m}^{-2}\,\text{d}^{-1}$) by multiplying the depth of the first soil layer (0.5 m); Ø is the soil porosity.

2.2 Input data preparation, model initialization and simulation

We developed gridded (32 km × 32 km), 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 US, 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-h 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 in 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), US 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.

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 $< 1\ \text{g m}^{-2}$ for carbon storage, $< 0.1\ \text{g m}^{-2}$ 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

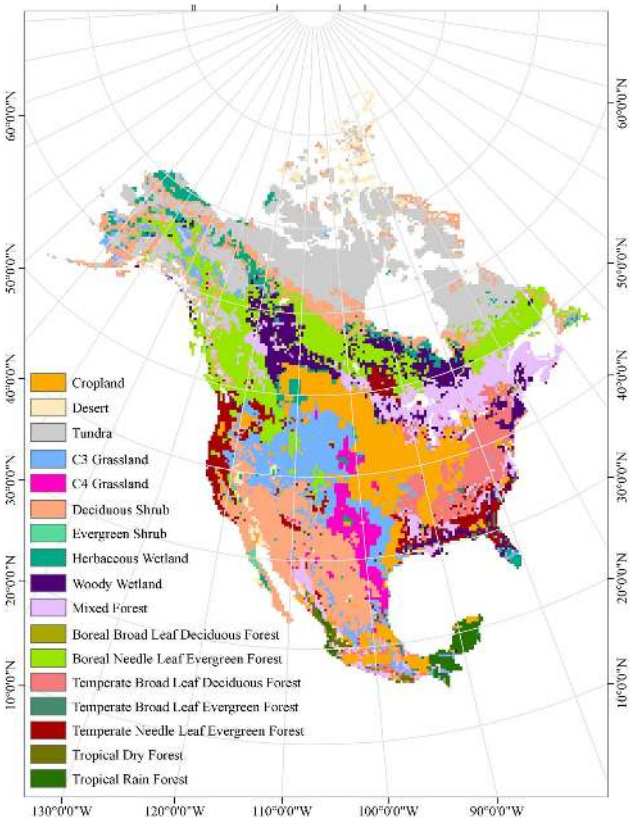

Fig. 4. Contemporary vegetation map used in this study (The year 2000 is shown).

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).

2.4 Model verification

Two forest sites (Durham forest and Hubbard Brook forest) and two wetland sites (Alaska wetland and Sallie's fen) 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 US 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's fen (P. 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–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 Sallie's fen (Fig. 5c), and underestimated CH_4 flux at Alaskan wetland site (Fig. 5b). 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's 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).

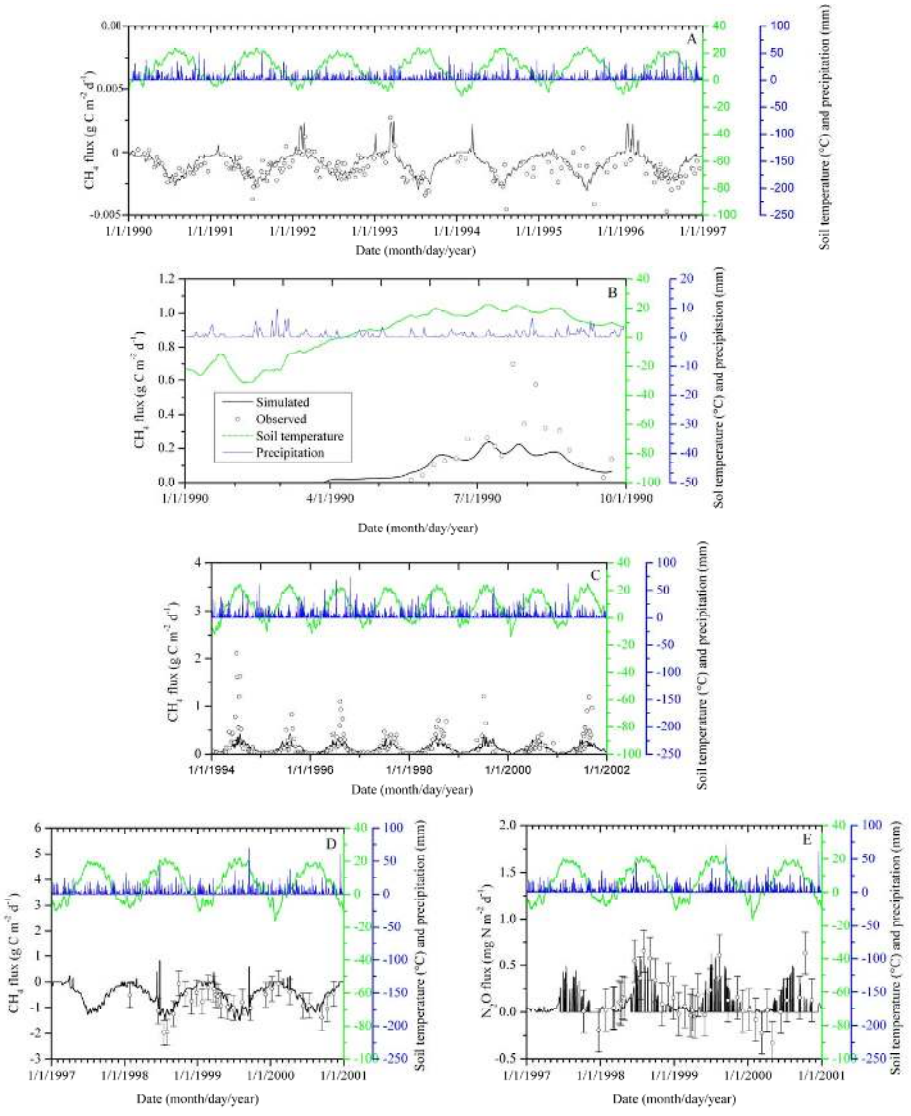

Fig. 5. Comparison of the DLEM-estimated CH_4 and N_2O fluxes with field observations – **(A)** CH_4 flux in Durham Forest (42° N, 73° W) (TRAGNET); **(B)** CH_4 flux in Alaska wetland (64.8° N, 147.7° W) (TRAGNET); **(C)** CH_4 flux in Sallie's fen (43.21° N, 71.05° W) ; **(D)** CH_4 flux in Hubbard Brook Forest (43.95° N, 71.74° W) (Groffman et al., 2006, 2009); **(E)** N_2O flux in Hubbard Brook Forest (43.95° N, 71.74° W) (Groffman et al., 2006, 2009). The error bars in Fig. 4d and e 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)

Although the reliability of simulated CH_4 and N_2O fluxes was shown as the comparison with observational data (Fig. 5), inconsistencies exist, i.e. the underestimation of CH_4 emission in Sallie's fen and simulated peak in CH_4 flux that were rarely shown in observational data for Durham forest (Fig. 5). For the simulated CH_4 peak while rarely shown in observations, it might due to the low sampling frequency of field work. The underestimation of CH_4 emission in Sallie's fen might be due to either the external environmental trigger which are not included in the model (Tokida et al., 2007; Shoemaker and Schrag 2010), or the contribution to CH_4 from the microbial mat systems during summer season (Shoemaker and Schrag, 2010).

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
$V_{CH_4ProMax}$ ($g\,C\,m^{-3}\,day^{-1}$)	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
$V_{CH_4OxidairMax}$ ($g\,C\,m^{-3}\,day^{-1}$)		0.10	< 0.001-103.7	Sitaula et al., 1995; Segers, 1998; Saari et al., 2004
$V_{CH_4OxidtransMax}$ ($g\,C\,m^{-3}\,day^{-1}$)		0.5	0->51.84	Segers, 1998
$V_{CH_4Oxidsoilmax}$ ($g\,C\,m^{-3}\,day^{-1}$)		0.5	0->51.84	Segers, 1998
Km_{CH_4Prod} ($g\,C\,m^{-3}$)		10	1.68–91.2	Lokshina et al., 2001
$Km_{CH_4Oxidair}$ (ppm)		10	5–18	Nedwell and Watson, 1995; Arah and Stephen, 1998; Saari et al., 2004
$Km_{CH_4Oxidtrans}$ ($g\,C\,m^{-3}$)		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
$Km_{CH_4Oxidsoil}$ ($g\,C\,m^{-3}$)		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 (2009) study. ** Assume cropland has higher half-saturation coefficient for nitrification and denitrification than natural ecosystems.

3 Results and analyses

3.1 Temporal patterns of CH_4 and N_2O fluxes in North America

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 $0.01\,T\,g\,C\,a^{-1}$ over

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.

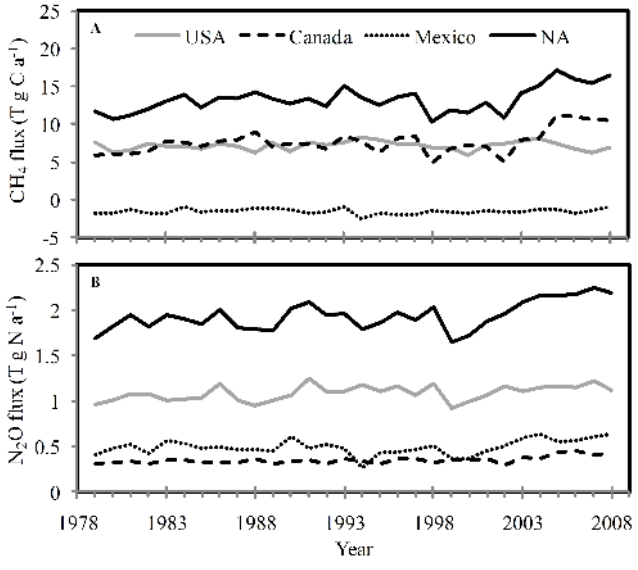

Fig. 6. CH_4 and N_2O fluxes in North America's terrestrial ecosystems by countries during 1979–2008.

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 US 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

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** forest	Temperate broad leaf deciduous	46.6° N, 128.47° E	Xiao et al., 2004
Changbaishan** forest	Temperate broad leaf evergreen	46.6° N, 128.47° E	Xiao et al., 2004
Gongga Mountain** forest	Temperate needle leaf evergreen	29.0 ∼ 30.33° N, 101.5 ∼ 102.25° E	Dong et al., 2003
Rondnia*,**	Tropical dry forest	10.5° S, 62.5° W	Garcia-Montiel et al., 2002
Rondnia*,**	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.

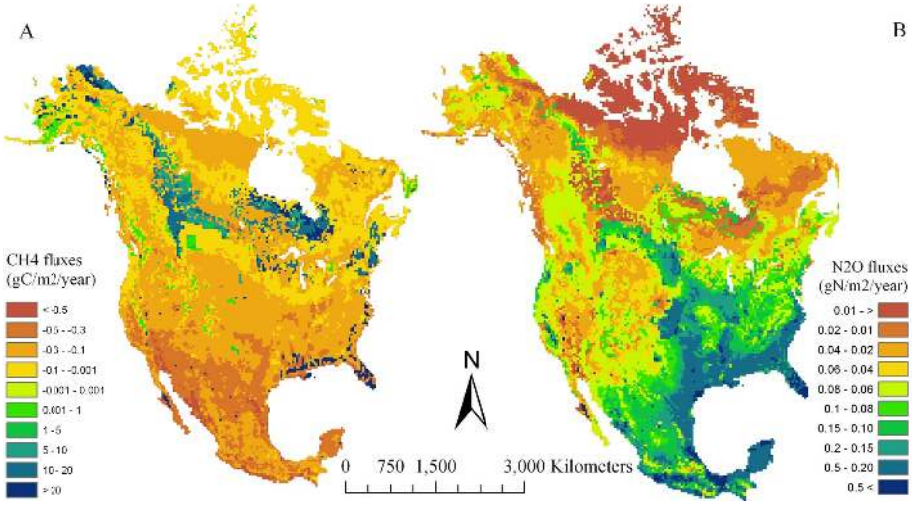

Fig. 7. Spatial distribution of **(A)** CH_4 and **(B)** N_2O fluxes in North America's terrestrial ecosystems during 1979–2008.

America where N_2O was released at a rate of approximately $0.01\,g\,N\,m^{-2}\,a^{-1}$.

3.3 CH_4 and N_2O fluxes in different countries

The US, 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 $7.16 \pm 0.58\,T\,g\,C\,a^{-1}$ for the US, $7.68 \pm 1.59\,T\,g\,C\,a^{-1}$ for Canada, and $-0.15 \pm 0.03\,T\,g\,C\,a^{-1}$ for Mexico. The US 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

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

Major ecosystem type	$V_{CH_4ProMax}$ ($g\,C\,m^{-3}\,day^{-1}$)	$V_{CH_4OxidairMax}$ ($g\,C\,m^{-3}\,day^{-1}$)	$V_{CH_4Oxidtrans}$ ($g\,C\,m^{-3}\,day^{-1}$)	$V_{CH_4Oxidsoilmax}$ ($g\,C\,m^{-3}\,day^{-1}$)	Km_{CH_4Prod} ($g\,C\,m^{-3}$)	$Km_{CH_4Oxidair}$ (ppm)	$Km_{CH_4Oxidtrans}$ ($g\,C\,m^{-3}$)	$Km_{CH_4Oxidsoil}$ ($g\,C\,m^{-3}$)
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

$1.09 \pm 0.08\,T\,g\,N\,a^{-1}$ for the US, $0.35 \pm 0.04\,T\,g\,N\,a^{-1}$ for Canada, and $0.50 \pm 0.08\,T\,g\,N\,a^{-1}$ for Mexico. The US, 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 $5.7\,G\,g\,C\,a^{-1}$ ($1\,G\,g = 10^9\,g$) in the US and $91.7\,G\,g\,C\,a^{-1}$ in Canada, while CH_4 consumption increased $0.2\,G\,g\,C\,a^{-1}$

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.

in Mexico. Our results also indicate that N_2O emission increased at rates of $4.2\,G\,g\,N\,a^{-1}$ in the US, $2.9\,G\,g\,N\,a^{-1}$ 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 dominate CH_4 emissions in North America with a source of $17.75 \pm 1.63\,T\,g\,C\,a^{-1}$ (Table 6). All the other ecosystems acted as sinks for atmospheric CH_4 with a total sink of $3.06 \pm 0.14\,g\,C\,a^{-1}$, of which forest and shrub contributed 36.93% and 20.92%, respectively. All ecosystems functioned as sources of N_2O. Forest, grassland, shrub, and cropland contributed 32.21%, 11.24%, 12.72%, and 28.82%, 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.

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 $32\,km \times 32\,km$ 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 US 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 ($17.75 \pm 1.63\,T\,g\,C\,a^{-1}$). 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 across the North America for the period of interest. In contrast, 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 up-to-date and detailed input data, our modeling approach addressed various ecosystem processes and multiple

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 US	4.13		Potter et al., 2006
DLEM	1996–2005	Wetlands in the continental US	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 US		0.50–0.74	Li et al., 1996
DLEM	1990	Cropland in the continental US		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.

environmental factors that control CH_4 and N_2O fluxes in terrestrial ecosystems.

There are only a few studies of N_2O at large spatial and temporal 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 $1.85 \pm 0.11\,T\,g\,N\,a^{-1}$, comparable to $2.08 \pm 0.05\,T\,g\,N\,a^{-1}$ 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 US is higher than Del Grosso et al's estimate for major crops (2005), but is lower than Li et al's estimate for cropland in the continental US (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 $9 \pm 4.5\,T\,g\,C\,a^{-1}$ (Chen and Prinn, 2006). However, their estimate did not consider the CH_4 flux from lower latitude, i.e. Mexico.

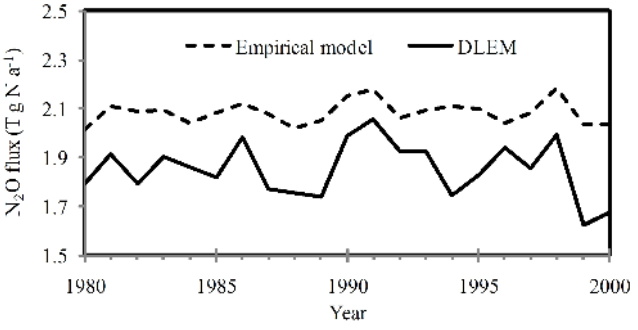

Fig. 8. Comparison of the DLEM-derived N_2O fluxes with the estimations by an empirical model (Xu et al., 2008). The regression model is: the DLEM-derived $N_2O = 0.8887 \times$ Empirically-modeled N_2O, $R^2 = 0.39$; $P < 0.01$.

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 $9.99 \pm 0.93\,g\,C\,m^{-2}\,a^{-1}$ and woody wetland at a rate of $7.87 \pm 0.81\,g\,C\,m^{-2}\,a^{-1}$. 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 $0.13 \pm 0.01\,g\,C\,m^{-2}\,a^{-1}$, which is close to Curry's (2007) estimate at $0.13\,g\,C\,m^{-2}\,a^{-1}$, and Ridgwell et al.'s (1999) estimate at $0.14\,g\,C\,m^{-2}\,a^{-1}$, yet is slightly lower than Dutaur and Verchot's (2007) estimate at $0.20\,g\,C\,m^{-2}\,a^{-1}$; the DLEM-estimated CH_4 uptake rate by grassland is $0.18 \pm 0.01\,g\,C\,m^{-2}\,a^{-1}$, which is consistent with Curry et al.'s (2007) estimate at $0.17\,g\,C\,m^{-2}\,a^{-1}$ and Dutaur and Vechot's (2007) estimate at $0.17\,g\,C\,m^{-2}\,a^{-1}$, yet

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		$-0.055 \sim -0.575$	-0.109	-0.112	-0.075
Boreal forest	-0.128 ± 0.010		$-0.074 \sim -0.430$	-0.125	-0.198	-0.140
Temperate forest	-0.178 ± 0.012		$-0.068 \sim -1.15$	-0.155	-0.428	-0.181
Tropical dry forest	-0.244 ± 0.013		$-0.082 \sim -0.520$	-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	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	Geometric: 6.075 ± 1.575				

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

is slightly lower than Ridgwell et al.'s (1999) estimate as $0.24\,g\,C\,m^{-2}\,a^{-1}$; the DLEM-estimated CH_4 uptake rate by cropland is $0.12 \pm 0.01\,g\,C\,m^{-2}\,a^{-1}$, 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 $0.018 \pm 0.002\,g\,N\,m^{-2}\,a^{-1}$ in this study, which is more than 50% higher than Potter et al.'s (1996) estimation at 0.003–$0.011\,g\,N\,m^{-2}\,a^{-1}$; 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 (1996) estimate (Table 9). The DLEM-estimated N_2O flux from desert is $0.015 \pm 0.003\,g\,N\,m^{-2}\,a^{-1}$, which is almost three times of Potter et al.'s (1996) estimate; 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 emission from wetland, and current state of lacking regional estimation of N_2O from wetland, it is highly needed to have further

efforts on this issue. Emission of N_2O from croplands in the US 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. 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 (Sect. 3.4). The highest increasing rate of N_2O emission from the US was probably caused by large amount of cropland, which was an major source for atmospheric N_2O (Li et al., 1996) (Sect. 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 Eq. 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.

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

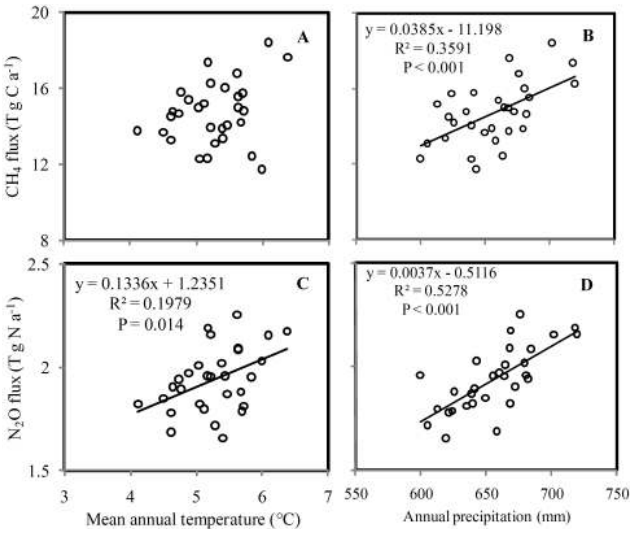

Fig. 9. Correlation between annual CH_4 and N_2O fluxes and mean annual temperature and annual precipitation – **(A)** Correlation between CH_4 flux and mean annual temperature; **(B)** Correlation between CH_4 flux and annual precipitation; **(C)** Correlation between N_2O flux and mean annual temperature;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

(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.

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; Ambus et al., 2006; Livesley et al., 2009). 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 DLEM model, 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} 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 US 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 data sets of wetland area and pasture management; the future incorporation of pasture into the DLEM simulation might be another way to reduce uncertainty of regional estimation of N_2O flux. 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.

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