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# Spatial and temporal patterns of CH<sub>4</sub> and N<sub>2</sub>O fluxes in terrestrial ecosystems of North America during 1979–2008: application of a global biogeochemistry model

#### H. Tian, X. Xu, M. Liu, W. Ren, C. Zhang, G. Chen, and C. Lu

Ecosystem Dynamics and Global Ecology (EDGE) Laboratory, School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL, 36849, USA

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

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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 biogeo-5 chemical 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  $14.69 \pm 1.64 \text{ Tg} \text{ Ca}^{-1}$  ( $1 \text{ Tg} = 10^{12} \text{ g}$ ) of CH<sub>4</sub>, and  $1.94\pm0.16$  T g N a<sup>-1</sup> of N<sub>2</sub>O were released from terrestrial ecosystems in North America. At the country level, both the United States and Canada acted as 10  $CH_4$  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<sub>2</sub>O emission in North America, the United States, Canada, and Mexico contributed 56.19%, 18.23%, and 25.58%, respectively, 15

- 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, flux, both mean annual temperature and annual precipitation were
- $_{20}$  fect on annual CH<sub>4</sub> flux, both mean annual temperature and annual precipitation were significantly correlated to annual N<sub>2</sub>O flux. The regional estimates and spatiotemporal patterns of terrestrial ecosystem CH<sub>4</sub> and N<sub>2</sub>O fluxes in North America generated in this study provide useful information for global change research and policy making.

#### 1 Introduction

<sup>25</sup> Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) are two potent greenhouse gases which in sum contribute to more than one fourth of global warming (Forster et al., 2007). Although

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the concentrations of CH<sub>4</sub> and N<sub>2</sub>O in the atmosphere are relatively low, their warming potentials are super-high compared to carbon dioxide (Denman et al., 2007). CH<sub>4</sub> and N<sub>2</sub>O also play significant roles in ozone layer chemistry (Denman et al., 2007; Forster et al., 2007). Similar to the increase of atmospheric CO<sub>2</sub> concentration, the concentrations of these two gases increased at rates of 1% a<sup>-1</sup> for CH<sub>4</sub> and 0.25% a<sup>-1</sup> for N<sub>2</sub>O with significant inter-annual fluctuations (Forster et al., 2007; Tueut et al., 2007; Rigby et al., 2008). Although the importance of CH<sub>4</sub> and N<sub>2</sub>O emissions in changing the Earth's climate has been recognized, scientific community has placed large emphasis on the CO<sub>2</sub> problem. Understanding and quantifying CH<sub>4</sub> and N<sub>2</sub>O 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<sub>4</sub> and N<sub>2</sub>O, 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 ter-15 restrial 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 (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  $CH_4$ 20 and  $N_2O$  fluxes over terrestrial ecosystems in North America (Bridgham et al., 2006). For example, Zhuang et al. estimated that soils in Canada and Alaska emitted 7.1 and  $3.8 \text{ Tg} \text{ CH}_4 \text{ a}^{-1}$ , respectively, during the 1990s (Zhuang et al., 2004). Bridgham et al. estimated that  $CH_4$  emission in North America's wetlands is  $9TgCH_4a^{-1}$  (Bridgham et al., 2006). Using a satellite-derived modeling approach, Potter et al. estimated 25 that the CH<sub>4</sub> emission from the natural wetlands in the conterminous United States is 5.5 T g CH<sub>4</sub> a<sup>-1</sup> (Potter et al., 2006). Several studies also reported the fluxes of N<sub>2</sub>O in terrestrial ecosystems at global and regional scales using empirical approaches (Xu et al., 2008). While these studies improved our understanding of CH<sub>4</sub> and N<sub>2</sub>O fluxes

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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;

<sup>5</sup> Phillips et al., 2001a, b), 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., 2004, 2007). Large uncertainty still exists in the magnitudes, spatial and temporal patterns of CH<sub>4</sub> and N<sub>2</sub>O 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 cli-

- <sup>15</sup> water in the land ecosystems. The DLEM considers multiple factors including climate, atmospheric compositions (CO<sub>2</sub>, O<sub>3</sub>), precipitation chemistry (nitrogen composition), natural disturbances (fire, insect/disease, hurricane, etc.), land-use/land-cover change, and land management (harvest, rotation, fertilization, irrigation, etc.)(Zhang et al., 2007; Zhang, 2008; Tian et al., 2005, 2008, 2010; Ren et al., 2007a, b; Ren, 2009; Lv, 2009; Liu et al., 2008; Chen et al., 2006). 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, b; Ren, 2009; Lv, 2009; Liu et al., 2008; Chen et al., 2006) and USA (Zhang et al., 2007; Zhang, 2008; Tian et al., 2008, 2010).

In this study, we enhanced the model's capability by addressing the biogeochemical processes of CH<sub>4</sub> and N<sub>2</sub>O and simulated CH<sub>4</sub> and N<sub>2</sub>O fluxes over terrestrial ecosystems in North America from 1979 to 2008. The objectives of this study are: 1) to enhance a process-based ecosystem model to simulate CH<sub>4</sub> and N<sub>2</sub>O fluxes; 2) to compare modeled results with field observations and other regional estimates; 3) to estimate CH<sub>4</sub> and N<sub>2</sub>O fluxes in North America's terrestrial ecosystems from 1979 to

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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, 5 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 10 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, 15 including the 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 redis-
- tribution 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 distributions. 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

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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; plant physiology component yields influences on biophysics compo-

- nent through changes in leaf area index (LAI), canopy conductance, and transpiration, on soil biogeochemistry component through litter-fall, and on dynamic vegetation component through biomass growth; dynamic vegetation component yields influences on plant physiology and soil biogeochemistry components through shifts of plant function type (PFT); soil biogeochemistry component yields influences on dynamics vegetation and plant physiology accomponents through putrient flow: disturbances, land use
- tion 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

<sup>15</sup> including ecosystem carbon and nitrogen pools and fluxes (e.g. greenhouse gases) will enter atmosphere; and the water output and associated nutrient 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 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 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 varied spatial resolutions, from meters to kilometers, from regional to global. The additional information on the processes,

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interactions and feedbacks in the DLEM and associated input/output data (Fig. 1) can be found in our previous studies (Zhang et al., 2007; Zhang, 2008; Tian et al., 2005, 2008, 2010; Ren et al., 2007a, b; Ren, 2009; Lv, 2009; Liu et al., 2008; Chen et al., 2006).

<sup>5</sup> In this paper, we provided a detailed description of the CH<sub>4</sub> and N<sub>2</sub>O modules with an emphasis on major processes that control fluxes of CH<sub>4</sub> and N<sub>2</sub>O in terrestrial ecosystems (Fig. 2).

#### **2.1.1** The $CH_4$ module

The CH<sub>4</sub> exchanges between ecosystems and the atmosphere are a combination of CH<sub>4</sub> production, oxidation, and transportation from soil/water to the atmosphere. The 10 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 either is used as substrate for methane or enters the 15 atmosphere as  $CO_2$  via decomposition.  $CH_4$  oxidation, including the oxidation during CH<sub>4</sub> transport to the atmosphere, CH<sub>4</sub> 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<sub>4</sub>-related biogeochemical reactions in the DLEM are described by using the Michaelis-Menten equation 20 with two coefficients: maximum reaction rate and half saturation coefficient. Three pathways for CH<sub>4</sub> 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<sub>4</sub> flux is determined by the following equation: 25

$$F_{CH_4} = (F_P + F_D + F_E - F_{airoxid} - F_{oxidtrans}) \cdot H$$

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(1)

where  $F_{CH_4}$  is the flux of CH<sub>4</sub> between soil and the atmosphere (g C m<sup>-2</sup> d<sup>-1</sup>);  $F_P$  is plant-mediated transport from soil/water to the atmosphere (g C m<sup>-3</sup> d<sup>-1</sup>);  $F_D$  is the diffusive flux of CH<sub>4</sub> from water surface to the atmosphere (g C m<sup>-3</sup> d<sup>-1</sup>);  $F_E$  is the ebullitive CH<sub>4</sub> emission to the atmosphere;  $F_{airoxid}$  is the rate of atmospheric methane oxidation (g C m<sup>-3</sup> d<sup>-1</sup>);  $F_{oxidtrans}$  is the oxidized CH<sub>4</sub> during plant-mediated transport (g C m<sup>-3</sup> d<sup>-1</sup>); H is the soil depth of the first layer which is assumed to be the only habitat for soil methanogenesis and methanotrophy (0.5 m in this study).

#### CH<sub>4</sub> production

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

$$CH_{4_{\text{prod}}} = V_{CH_{4_{\text{prod}} \text{max}}} \cdot \frac{[DOC]}{[DOC] + K m_{CH_{4_{\text{prod}}}}} \cdot f(T_{\text{soil}}) f(\text{pH}) f_{\text{prod}}(W)$$
(2)

$$\frac{d[CH_4]}{dt} = CH_{4_{\text{prod}}} - F_{\text{P}} - F_{\text{D}} - F_{\text{E}} - CH_{4_{\text{oxidsoil}}}$$
(3)

where  $CH_{4_{prod}}$  is the production of  $CH_4$  in soil/water ( $gCm^{-3}d^{-1}$ );  $V_{CH_{4_{prod}max}}$  is the maximum rate of  $CH_4$  production ( $gCm^{-3}d^{-1}$ ), [DOC] is the concentration of DOC ( $gCm^{-3}$ );  $Km_{CH_{4_{prod}}}$  is the half-saturation coefficient of  $CH_4$  production ( $gCm^{-3}$ );  $f(T_{soil})$  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  $CH_4$  production is a multiplier that describes the effect of soil pH on  $CH_4$  production  $CH_4$  production;  $f_{prod}(W)$  is a multiplier that describes the effect of soil moisture on  $CH_4$  production;  $[CH_4]$  is the concentration of  $CH_4$  in water ( $gCm^{-3}$ );  $CH_{4_{oxidsoil}}$  is the oxidation rate of  $CH_4$  in soil/water ( $gCm^{-3}d^{-1}$ ).

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#### CH<sub>4</sub> 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/water, mainly simulates the oxidation of atmospheric  $CH_4$  in the soil/water; 2) the process of  $CH_4$  oxidation in the soil/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/water to the atmosphere. The DLEM assumes that the process of  $CH_4$  oxidation in soil/water includes the  $CH_4$  oxidation during ebullition and diffusion because these two processes only

10 occur in water.

5

#### Atmospheric CH<sub>4</sub> oxidation

Oxidation of atmospheric CH<sub>4</sub> is estimated as:

$$F_{\text{airoxid}} = V_{\text{OxidairMax}} \cdot \frac{[\text{AtmCH}_4]}{[\text{AtmCH}_4] + K m_{\text{CH}_{4_{\text{oxida}}}}} \cdot f(T_{\text{soil}}) \cdot f(\text{pH}) \cdot f_{\text{oxid}}(W)$$
(4)

where  $V_{\text{OxidairMax}}$  is the maximum oxidation rate of atmospheric CH<sub>4</sub> (g C m<sup>-3</sup> d<sup>-1</sup>);, <sup>15</sup>  $Km_{\text{CH}_{4_{\text{oxida}}}}$  is the half saturation coefficient of atmospheric CH<sub>4</sub> oxidation (g C m<sup>-3</sup>); [AtmCH<sub>4</sub>] is the atmospheric CH<sub>4</sub> concentration (g C m<sup>-3</sup>);  $f_{\text{oxid}}(W)$  is a multiplier that describes the effect of soil moisture on atmospheric CH<sub>4</sub> oxidation. Because the atmospheric CH<sub>4</sub> oxidation is mainly carried out by soil methanotrophy, and low soil organic matter means lower soil microbial biomass (Conrad, 1996), the DLEM assumes that <sup>20</sup> there is no atmospheric CH<sub>4</sub> oxidation when soil organic matter is less than 10 g C m<sup>-2</sup>.

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#### CH<sub>4</sub> 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:

$$CH_{4_{\text{oxidtrans}}} = \min(V_{\text{OxidtransMax}} \cdot \frac{F_{\text{P}}}{F_{\text{P}} + Km_{\text{CH}_{4_{\text{oxidtrans}}}}} \cdot f(T_{\text{air}}), F_{\text{P}})$$
(5)

<sup>5</sup> where  $CH_{4_{oxidtrans}}$  is the oxidation rate of  $CH_4$  during plant-mediated transport  $(g C m^{-3} d^{-1})$ ; min() represents a function which returns the smaller one of two number in the brackets;  $V_{OxidtransMax}$  is the maximum rate of  $CH_4$  oxidation  $(g C m^{-3} d^{-1})$ ;  $Km_{CH_{4_{oxidtrans}}}$  is the half saturation coefficient of soil  $CH_4$  oxidation during transportation  $(g C m^{-3})$ ;  $T_{air}$  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.

#### Soil/water CH<sub>4</sub> oxidation

The accumulated CH<sub>4</sub> in soil/water is oxidized at the rate of:

$$CH_{4_{\text{oxidsoil}}} = \min\left(V_{\text{OxidsoilMax}} \cdot \frac{[CH_4]}{[CH_4] + K m_{CH_{4_{\text{oxidsoil}}}}} \cdot f(T_{\text{soil}}) \cdot f(pH) \cdot f_{\text{oxid}}(W), [CH_4]\right)$$
(6)

where  $V_{\text{OxidsoilMax}}$  and  $Km_{\text{CH}_{4_{\text{oxidsoil}}}}$  are maximum soil/water CH<sub>4</sub> oxidation rate 15 (g C m<sup>-3</sup> d<sup>-1</sup>) and half saturation coefficient of CH<sub>4</sub> oxidation in soil/water (g C m<sup>-3</sup>), respectively; [CH<sub>4</sub>] is the concentration of CH<sub>4</sub> in soil/water (g C m<sup>-3</sup>).

#### CH<sub>4</sub> transport

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

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#### Ebullition

The ebullition transport of CH<sub>4</sub> from water to the atmosphere is estimated as:

 $F_{\rm E} = \max(([{\rm CH}_4] - 6),)$ 

where  $F_E$  is the flux of CH<sub>4</sub> from water to the atmosphere via ebullition (g C m<sup>-3</sup> d<sup>-1</sup>); max() represents a function which returns the larger one of two numbers in the brackets; 6 is the threshold value above which the dissolved CH<sub>4</sub> will form bubbles and leave water (g C m<sup>-3</sup>), and is equals to 0.5 mol CH<sub>4</sub> m<sup>-3</sup> (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<sub>4</sub> above this threshold value will leave water via bubbles in one day.

#### Plant-mediated transport

The plant-mediated CH<sub>4</sub> emission from water to the atmosphere is estimated as:

$$F_{\rm P} = V_{\rm trans, plant} \cdot \left( [\rm CH_4] - [\rm CH_4]_{\rm max} \right) \cdot \min\left( \frac{\rm GPP_{\rm day}}{\rm GPP_{\rm max}}, 1 \right)$$
(8)

 $[CH_4]_{max} = [AirCH_4] \cdot \beta$ 

<sup>15</sup> where  $F_P$  is the CH<sub>4</sub> transport via vascular plant (g C m<sup>-3</sup> d<sup>-1</sup>);  $V_{trans,plant}$  is the transport coefficient of CH<sub>4</sub> transportation through plant (d<sup>-1</sup>), which is set as 0.68 (Kettunen, 2003); [CH<sub>4</sub>]<sub>max</sub> is the maximum CH<sub>4</sub> concentration in soil solution (g C m<sup>-3</sup>); GPP<sub>day</sub> is the gross primary productivity of current day (g C m<sup>-2</sup> d<sup>-1</sup>); GPP<sub>max</sub> is the maximum daily GPP (g C m<sup>-2</sup> d<sup>-1</sup>), which is set as 5 in this study; [AirCH<sub>4</sub>] is the atmospheric <sup>20</sup> concentration of CH<sub>4</sub>;  $\beta$  is the Bunsen solubility coefficient (0.035 ml ml<sup>-1</sup>) (Yamamoto et al., 1976). Since there is no report on the plant-mediated transport of CH<sub>4</sub> 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.

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(7)

(9)

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#### 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 <sup>5</sup> atmosphere.

 $F_{\rm D} = V_{\rm exchange} \cdot ([CH_4] - [CH_4]_{\rm max})$ 

(10)

(11)

where  $V_{\text{exhange}}$  is the exchange coefficient of CH<sub>4</sub> through the interface of soil/water and the atmosphere (m d<sup>-1</sup>); it is set as 0.3 m d<sup>-1</sup> (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<sub>4</sub> production, consumption, and transport (Cao et al., 1995; Huang et al., 1998; Mer and Roger, 2001; Zhuang et al., 2004).
- <sup>15</sup> 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<sub>4</sub> 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<sub>4</sub> 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).

 $f(pH) = \begin{cases} 0.0 & pH \le 4.0 \text{ or } pH \ge 10.0\\ \frac{1.02}{1+1\ 000\ 000 \cdot exp^{(-2.5 \cdot pH)}} & 4.0 < pH \le 7.0\\ \frac{1.02}{1+1\ 000\ 000 \cdot exp^{(-2.5 \cdot (14.0 - pH))}} & 7.0 < pH < 10.0 \end{cases}$ 

where pH is the pH value of the soil profile.

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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.

$${}_{5} f(T) = \begin{cases} 0.0 & T < -5.0\\ Q_{10}^{\frac{T-30}{10}} & 30 > T \ge -5.0\\ 1 & T \ge 30 \end{cases}$$
(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.

$$f_{\text{prod}}(W) = \begin{cases} 0 & \text{for } \text{vwc} \leq \text{vwc}_{\text{fc}} \\ \left(\frac{\text{vwc} - \text{vwc}_{\text{fc}}}{\text{vwc}_{\text{sat}} - \text{vwc}_{\text{fc}}}\right)^2 \cdot 0.368 \cdot e^{\left(\frac{\text{vwc} - \text{vwc}_{\text{fc}}}{\text{vwc}_{\text{sat}} - \text{vwc}_{\text{fc}}}\right)} & \text{for } \text{vwc} \geq \text{vwc} > \text{vwc}_{\text{fc}} \\ 1 & \text{for } \text{vwc} > \text{vwc}_{\text{sat}} \end{cases}$$

$$f_{\text{oxid}}(W) = 1 - f_{\text{prod}}(W) \qquad (13)$$

where vwc is the volumetric water content of the top soil layer; vwc<sub>fc</sub> is the field capacity and vwc<sub>sat</sub> 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.

#### 15 2.1.2 The N<sub>2</sub>O 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.

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#### 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).

$$\begin{split} N_{\text{nitrif}} &= \min\left(N_{\text{pot,nitrif}}, N_{\text{NH}_4}\right) \\ N_{\text{pot,nitrif}} &= V_{\text{max,nit}} \cdot \frac{N_{\text{NH}_4}}{N_{NH_4} + K_{\text{nit}}} \cdot f_{\text{nit}}(T_{\text{soil}}) \cdot f_{\text{nit}}(W) \\ f_{\text{nit}}(T_{\text{soil}}) &= Q_{10.\text{nit}}^{\left(\frac{T_{\text{soil}} - T_{\text{opt,nit}}}{10}\right)} \\ f_{\text{nit}}(W) &= \begin{cases} 1.17 \cdot \frac{\text{vwc}}{\text{vwc}_{\text{fc}}} + 0.165 & \text{for vwc} < \text{vwc}_{\text{fc}} \\ 1 - 0.1 \cdot \frac{\text{vwc}}{\text{vwc}_{\text{fc}}} & \text{for vwc} \ge \text{vwc}_{\text{fc}} \end{cases} \end{split}$$

<sup>5</sup> where N<sub>nitrif</sub> is the nitrification rate (g N m<sup>-3</sup> d<sup>-1</sup>); N<sub>pot,nitrif</sub> is the potential nitrification rate (g N m<sup>-3</sup> d<sup>-1</sup>); N<sub>NH4</sub> is the concentration of NH<sup>+</sup><sub>4</sub> in the soil (g N m<sup>-3</sup>); V<sub>max,nit</sub> is a parameter describing potential nitrification rate without limitation (g N m<sup>-3</sup> d<sup>-1</sup>); K<sub>nit</sub> is the half-saturation concentration of soil NH<sup>+</sup><sub>4</sub> for the maximum nitrification rate (g N m<sup>-3</sup>); f<sub>nit</sub>(T<sub>soil</sub>) is a multiplier that describes the effect of soil temperature on nitrification; T<sub>soil</sub> is the soil temperature (°C); f<sub>nit</sub>(W) is a multiplier that describes the effect of water content on nitrification (Lin et al., 2000; Riedo et al., 1998); Q<sub>10,nit</sub> is the temperature for nitrification, which is set as 2; T<sub>opt,nit</sub> 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<sub>fc</sub> is the soil field capacity.

#### 15 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

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et al., 2000).

$$\begin{split} N_{\text{denitrif}} &= \min\left(N_{\text{pot,denitrif}}, N_{\text{NO}_3}\right) \\ N_{\text{pot,denitrif}} &= V_{\text{max,denit}} \cdot \frac{N_{\text{NO}_3}}{N_{\text{NO}_3} + K_{\text{denit}}} \cdot f_{\text{denit}}(T_{\text{soil}}) \cdot f_{\text{denit}}(W) \\ f_{\text{denit}}(T_{\text{soil}}) &= Q_{10.\text{denit}}^{\left(\frac{T_{\text{soil}} - T_{\text{opt,denit}}}{10}\right)} \\ f_{\text{denit}}(W) &= \begin{cases} 0.0 & \text{for vwc} < \text{vwc}_{\text{fc}} \\ \frac{\text{vwc}}{\text{vwc}_{\text{fc}}} & \text{for vwc} \ge \text{vwc}_{\text{fc}} \end{cases} \end{split}$$

where  $N_{\text{denitrif}}$  is the denitrification rate (g N m<sup>-3</sup> d<sup>-1</sup>);  $N_{\text{pot,denitrif}}$  is the potential nitrification rate (g N m<sup>-3</sup> d<sup>-1</sup>);  $N_{\text{NO}_3}$  is the concentration of NO<sub>3</sub><sup>-</sup> in the soil (g N m<sup>-3</sup>);  $V_{\text{max,denit}}$ 

<sup>5</sup> is a parameter describing potential denitrification rate without limitation (g N m<sup>-3</sup> d<sup>-1</sup>); K<sub>denit</sub> is the half-saturation concentration of soil NO<sub>3</sub><sup>-1</sup> for the maximum denitrification rate (g N m<sup>-3</sup>); f<sub>denit</sub>(T<sub>soil</sub>) is a multiplier that describes the effect of soil temperature on denitrification; f<sub>denit</sub>(W) is a multiplier that describes the effect of water content on denitrification (Lin et al., 2000; Riedo et al., 1998); Q<sub>10,denit</sub> is the temperature sensitivity of denitrification, which is set as 3; and T<sub>opt,denit</sub> is the optimum temperature for denitrification, which is set as 25 °C following Lin et al. (2000).

#### N<sub>2</sub>O emission

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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  $N_2$ ).

$$F_{N_2O} = (0.001 \cdot N_{\text{nitrif}} + N_{\text{denitrif}}) \cdot \frac{10^{\text{WFPS} \cdot 0.026 - 1.66}}{(1 + 10^{\text{WFPS} \cdot 0.026 - 1.66})}$$
(16)

where  $F_{N_2O}$  is the fluxes of N<sub>2</sub>O from soil to the atmosphere (g N m<sup>-3</sup> d<sup>-1</sup>), 0.001 is the proportion of nitrification product released as gaseous nitrogen (Lin et al., 2000), and it 2845

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is converted to fluxes in the unit area  $(gNm^{-2}d^{-1})$  by multiplying the depth of the first soil layer (0.5 m); WFPS is the water filled pore spaces.

#### 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 United States, and Mexico). The climate dataset was generated based on North American Regional Reanalysis (NARR) dataset. 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 1900–15 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 (www.daac.ornl.gov). Fertilization data for North America was developed by combin-
- ing several data sources, including Food and Agriculture Organization (FAO) countrylevel 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 reprojected to a consistent projection system for driving the DLEM model. The annual atmospheric concentration of CO<sub>2</sub> before 1959 was estimated by VEMAP (The Vegeta-
- tion/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. 3) was developed using

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different sources of data, including global land-cover derived from Landsate 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).

The implementation of DLEM simulation includes the following runs: 1) equilibrium

- <sup>5</sup> 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<sub>2</sub> in the year 1900 to drive the model run to an equilibrium state (i.e. the inter-annual variations are <0.1 g m<sup>-2</sup> for carbon storage, <0.1 g m<sup>-2</sup> 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<sub>2</sub> concentration and nitrogen deposition inputs from 1901 to 2008 to simulate CH<sub>4</sub> and N<sub>2</sub>O 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<sub>4</sub> and N<sub>2</sub>O fluxes
- <sup>15</sup> 2008 were analyzed to show the spatial and temporal patterns of  $CH_4$  and  $N_2O$  huxes in North America's terrestrial ecosystems. Urban was treated as grassland, which is the same as in the other terrestrial biosphere model (McGuire et al., 2001).

#### 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 sitelevel 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 for the model simulation. We used measurement data of CH<sub>4</sub> and N<sub>2</sub>O 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<sub>4</sub> fluxes (Table 2), and 18 sites for N<sub>2</sub>O fluxes (Table 3). Finally, a suite of parameters (eight for CH<sub>4</sub> module, and four for N<sub>2</sub>O 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 not used in model parameterization were selected for site-level model verification (Fig. 4).

- 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 (P. Crill, personal communication, 2008). We retrieved the sitelevel, model-driven data from our regional dataset for model run because the input data
- at these sites were unavailable. Four simulations for CH<sub>4</sub> and one for N<sub>2</sub>O showed that model results are significantly correlated with observational data even though the DLEM model underestimated some fluxes (Fig. 4a–e). While the general seasonal patterns of CH<sub>4</sub> flux at these sites were consistent with the observations, the DLEM model missed a few CH<sub>4</sub> flux pulses during the peak growing season in the Salle's fen
- (Fig. 4c), and underestimated CH<sub>4</sub> flux at Alaskan wetland site (Fig. 4b). For the N<sub>2</sub>O flux, the DLEM model well captured the seasonal pattern and annual flux of N<sub>2</sub>O in Hubbard Brook forest, but missed several spikes in observational data (Fig. 4e). This phenomenon of peak fluxes in CH<sub>4</sub> and N<sub>2</sub>O 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. 4.



#### 3 Results and analyses

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#### 3.1 Temporal patterns of CH<sub>4</sub> and N<sub>2</sub>O fluxes in North America

The annual fluxes of CH<sub>4</sub> and N<sub>2</sub>O over the entire North America showed significant fluctuations during 1979–2008. The highest CH<sub>4</sub> emission was 18.42 T g C a<sup>-1</sup>
in 2005, and the lowest was 11.74 T g C a<sup>-1</sup> in 1998. Before 2001, the annual CH<sub>4</sub> flux was relatively constant with no obvious trend of change; however, since 2002 the CH<sub>4</sub> emission rate increased rapidly, reached the maximum in 2005, and decreased slightly since then (Fig. 5). The overall increasing rate of CH<sub>4</sub> flux was 0.10 T g C a<sup>-1</sup> over the past 30 years. The highest N<sub>2</sub>O emission was 2.25 T g N a<sup>-1</sup> in 2007, and the lowest was 1.66 T g N a<sup>-1</sup> in 1999 (Fig. 5). The overall increasing rate of N<sub>2</sub>O was 0.01T g C a<sup>-1</sup> 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<sup>-1</sup> for CH<sub>4</sub>, and 1.94±0.16 T g N a<sup>-1</sup> for N<sub>2</sub>O, respectively.

#### 3.2 Spatial distributions of CH<sub>4</sub> and N<sub>2</sub>O fluxes in North America

The CH<sub>4</sub> and N<sub>2</sub>O fluxes over the entire continent of North America showed significant spatial variations (Fig. 6). The spatial pattern of CH<sub>4</sub> 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<sub>4</sub>. A weak sink of CH<sub>4</sub> was also shown in the northeastern part of North America.

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

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 $0.01 \, g \, N \, m^{-2} \, a^{-1}$ .

years.

#### 3.3 CH<sub>4</sub> and N<sub>2</sub>O fluxes in different countries

The United States, Canada, and Mexico are three diverse countries with different landscapes and anthropogenic activities, resulting in various CH<sub>4</sub> and N<sub>2</sub>O flux scenarios. At the country level,  $CH_4$  flux was  $7.16\pm0.58$  T g C a<sup>-1</sup> for the United States,  $7.68 \pm 1.59 \text{ Tg Ca}^{-1}$  for Canada, and  $-0.15 \pm 0.03 \text{ Tg Ca}^{-1}$  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. 7). The country level N<sub>2</sub>O flux was  $1.09\pm0.08$  T g N a<sup>-1</sup> for the United States,  $0.35\pm0.04$  T g N a<sup>-1</sup> for Canada, and  $0.50\pm0.08$  T g N a<sup>-1</sup> for Mexico. The United States, Canada, and Mexico accounted for 10 56.19%, 18.23%, and 25.58%, respectively, of the continental emission of N<sub>2</sub>O (Fig. 7). 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  $5.7 \text{ Gg} \text{ Ca}^{-1}$  (1 Gg=10<sup>9</sup> g) in the United States and 91.7 GgCa<sup>-1</sup> in Canada, while  $CH_4$  consumption increased 0.2 G g C a<sup>-1</sup> in Mexico. Our results also 15 indicate that  $N_2O$  emission increased at rates of  $4.2 \text{ GgN a}^{-1}$  in the United States.  $2.9 \text{ Gg} \text{ Na}^{-1}$  in Canada and  $2.9 \text{ Gg} \text{ Na}^{-1}$  in Mexico, respectively, during the past 30

#### 3.4 CH<sub>4</sub> and N<sub>2</sub>O fluxes in different biomes

<sup>20</sup> CH<sub>4</sub> and N<sub>2</sub>O fluxes varied remarkably among different ecosystems. Due to the perennial or ephemeral inundated condition, wetlands dominant CH<sub>4</sub> emissions in North America with a source of 17.75±1.63 TgCa<sup>-1</sup>. All the other ecosystems acted as sinks for atmospheric CH<sub>4</sub> with a total sink of  $3.06\pm0.14$  gCa<sup>-1</sup>. Tundra, forest, grassland, shrub, cropland, and desert and others oxidized atmospheric CH<sub>4</sub> at rates <sup>25</sup> of  $0.41\pm0.03$  TgCa<sup>-1</sup>,  $1.13\pm0.07$  TgCa<sup>-1</sup>,  $0.47\pm0.03$  TgCa<sup>-1</sup>,  $0.64\pm0.02$  TgCa<sup>-1</sup>,

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 $0.32\pm0.03$  T g C a<sup>-1</sup>,  $0.10\pm0.01$  T g C a<sup>-1</sup>, respectively, accounting for -2.79% (negative means uptake), -7.67%, -3.19%, -4.34%, -2.17%, and -0.70% of the continental CH<sub>4</sub> emission (Table 6).

- All ecosystems functioned as sources of N<sub>2</sub>O. Tundra, forest, grassland, <sup>5</sup> shrub, wetland, cropland, and desert and others emitted  $0.07\pm0.01$  Tg N a<sup>-1</sup>,  $0.63\pm0.03$  Tg N a<sup>-1</sup>,  $0.22\pm0.04$  Tg N a<sup>-1</sup>,  $0.25\pm0.03$  Tg N a<sup>-1</sup>,  $0.19\pm0.01$  Tg N a<sup>-1</sup>,  $0.56\pm0.07$  Tg N a<sup>-1</sup>,  $0.03\pm0.004$  Tg N a<sup>-1</sup>, accounting for 3.68%, 32.21%, 11.24%, 12.72%, 9.78%, 28.82%, and 1.55%, respectively, for the N<sub>2</sub>O emission in North America's terrestrial ecosystems (Table 6).
- <sup>10</sup> The fluxes of CH<sub>4</sub> and N<sub>2</sub>O in each biome over the past 30 years varied significantly. From 1979 to 2008, the CH<sub>4</sub> emission increased at a rate of 103.9 G g C a<sup>-1</sup> in natural wetland, and the CH<sub>4</sub> oxidation increased at rates of 2.5 G g C a<sup>-1</sup> in forests, 0.8 G g C a<sup>-1</sup> in shrub, 0.8 G g C a<sup>-1</sup> in grassland, and 0.6 G g C a<sup>-1</sup> in desert and others, respectively. No significant changes were found for other ecosystem types. For <sup>15</sup> the same time period, the N<sub>2</sub>O emission increased at rates of 5.5 G g N a<sup>-1</sup> in cropland, 1.5 G g N a<sup>-1</sup> in grassland, 0.8 G g N a<sup>-1</sup> in tundra, and 0.3 G g N a<sup>-1</sup> in desert and others. We did not find significant changes for other ecosystem types.

#### 4 Discussion

#### 4.1 Regional comparison to other studies

<sup>20</sup> We estimated annual fluxes of  $CH_4$  and  $N_2O$  in terrestrial ecosystems of North America with a spatial resolution of 32 km×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<sup>-1</sup> (Denman et al., 2007), which is lower than its <sup>25</sup> areal portion of global natural wetland. This may be due to less tropical natural wet-



lands and rice paddies in this region, which are two strong emitters of CH<sub>4</sub> (Denman et al., 2007; Mitsch and Gosselink, 2007). While our regional estimations of CH<sub>4</sub> and N<sub>2</sub>O in North America's terrestrial ecosystems were comparable to previous studies, we found that there were differences at some specific areas or ecosystems. For ex-<sup>5</sup> ample, Zhuang et al (Zhuang et al., 2007) used a process-based model (Terrestrial Ecosystem Model) to estimate that the CH<sub>4</sub> emission in Alaska was 2.35 T g C a<sup>-1</sup> for the period of 1980–1996, which is 12% higher than our estimation of 2.10 T g C a<sup>-1</sup> for the same time period. However, their estimation of CH<sub>4</sub> emissions in Canada of 5.33 T g C a<sup>-1</sup> (Zhuang et al., 2004) is 26% lower than our estimation of 7.23 T g C a<sup>-1</sup>

- <sup>10</sup> 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  $4.13 \text{ Tg} \text{ Ca}^{-1}$ , which is 35% lower than our estimate of  $6.34 \text{ Tg} \text{ Ca}^{-1}$  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<sup>-1</sup>, which is only 38% of our estimation (17.75±1.63 T g C a<sup>-1</sup>). Bridgham et al. (2006) used site specific estimates of CH<sub>4</sub> 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 contract, the DLEM estimates account for spatial
- variability of wetlands, soils and climate that give rise to place to place differences in CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> and N<sub>2</sub>O fluxes in terrestrial ecosystems.

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There are only a few studies of N<sub>2</sub>O 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<sub>2</sub>O 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<sub>2</sub>O flux was  $1.85\pm0.11$  T g N a<sup>-1</sup>, comparable to  $2.08\pm0.05$  T g N a<sup>-1</sup> estimated by Xu et al. (2008). Inter-annual variations of N<sub>2</sub>O fluxes also showed a good agreement between these two studies ( $R^2$ =0.39; P<0.01). The spatial correlation of N<sub>2</sub>O 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<sub>2</sub>O 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 (Li et al., 1996) (Table 7). Putting our estimate at the global context, DLEM-estimated N<sub>2</sub>O flux from North America accounted for 20%
- <sup>15</sup> the global context, DLEM-estimated N<sub>2</sub>O flux from North America accounted for 20% of the global N<sub>2</sub>O source of  $9.4 \text{ Tg} \text{ Na}^{-1}$  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<sup>-1</sup> (Chen and Prinn, 2006). However, their estimate did not consider the  $CH_4$  flux from lower latitude. Also one study reported the anthropogenic emissions of  $CH_4$  and  $N_2O$  from North America (Kort et al., 2008). In their study, the anthropogenic emissions over North America were estimated at 36.75 T g C and 2.74 T g N in 2003 (Kort et al., 2008). Given that the natural flux accounts for 28–43% of total  $CH_4$ flux, and ~50% of total  $N_2O$  flux if removing oceanic contribution (Denman et al., 2007), DLEM-estimated  $CH_4$  and  $N_2O$  flux (15.75 T g C and 2.1 T g N in 2003) are comparable to the inverse results.

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#### 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<sub>4</sub> at a rate of  $9.99\pm0.93$  g C m<sup>-2</sup> a<sup>-1</sup> and woody wetland at a rate of  $7.87\pm0.81$  g C m<sup>-2</sup> a<sup>-1</sup>. The strongest sink resided in subtropical/tropical dry forest, 5 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<sub>4</sub> uptake rate by boreal forest is  $0.13\pm0.01$  g C m<sup>-2</sup> a<sup>-1</sup>, which is close to Curry's estimate at  $0.13 \text{ g Cm}^{-2} \text{ a}^{-1}$  (Curry, 2007), and Ridgwell et al.'s estimate at  $0.14 \text{ g Cm}^{-2} \text{ a}^{-1}$  (Ridgwell et al., 1999), yet is slightly lower than 10 Dutaur and Verchot's estimate at  $0.20 \,\mathrm{gC}\,\mathrm{m}^{-2}\,\mathrm{a}^{-1}$  (Dutaur and Verchot, 2007); the DLEM-estimated CH<sub>4</sub> uptake rate by grassland is  $0.18\pm0.01$  g C m<sup>-2</sup> a<sup>-1</sup>, which is consistent with Curry et al.'s estimate at  $0.17 \text{ g Cm}^{-2} \text{ a}^{-1}$  (Curry, 2007) and Dutaur and Vechot's estimate at  $0.17 \,\text{g}\,\text{C}\,\text{m}^{-2}\,\text{a}^{-1}$  (Dutaur and Verchot, 2007), yet is slightly lower than Ridgwell et al.'s estimate as  $0.24 \text{ g Cm}^{-2} \text{ a}^{-1}$  (Ridgwell et al., 1999); the DLEMestimated CH<sub>4</sub> uptake rate by cropland is  $0.12\pm0.01$  g C m<sup>-2</sup> a<sup>-1</sup>, which is close to 0.11-0.15 g C m<sup>-2</sup> a<sup>-1</sup> (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<sub>2</sub>O 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<sub>2</sub>O flux from tundra is estimated at 0.018±0.002 g N m<sup>-2</sup> a<sup>-1</sup> in this study, which is more than 50% higher than Potter et al.'s estimation at 0.003–0.011 g N m<sup>-2</sup> a<sup>-1</sup> (Potter et al., 1996); while it is in the lower end of 0.002–0.251 g N m<sup>-2</sup> a<sup>-1</sup> 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<sub>2</sub>O flux from tropical rain forest is identical to Potter et al.'s estimate (Table 9). The





DLEM-estimated N<sub>2</sub>O flux from desert is 0.015 ±0.003 g N m<sup>-2</sup> a<sup>-1</sup>, which is almost three times of Potter et al.'s estimate (Potter et al., 1996); however, it is still reasonable comparing with Guilbault and Matthias's filed observation of 0.04 g N m<sup>-2</sup> a<sup>-1</sup> at Sonoran Desert (Guilbault and Matthias, 1998). Our estimated N<sub>2</sub>O 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<sub>2</sub>O emission from wetland, and current state of lacking regional estimation of N<sub>2</sub>O from wetland, it is highly urgent to have further efforts on this issue. Emission of N<sub>2</sub>O from croplands in the United States in 1990 (0.187±0.139 g N m<sup>-2</sup> a<sup>-1</sup>) was also comparable to another
estimates of 0.186–0.204 g N m<sup>-2</sup> a<sup>-1</sup> by Li et al. (1996).

#### 4.3 Environmental controls on CH<sub>4</sub> and N<sub>2</sub>O fluxes

Inter-annual fluctuation of CH<sub>4</sub> and N<sub>2</sub>O 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); 15 this is consistent with our study, which shows that the droughts in 1994, 1999, and 2002 resulted in relative low N<sub>2</sub>O 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<sub>4</sub> and N<sub>2</sub>O fluxes (Steudler et al., 1989; Ding et al., 2004; Liu and Greaver, 2009), while rising 20 atmospheric CO<sub>2</sub> increased CH<sub>4</sub> emission (Hutchin et al., 1995) yet decreased N<sub>2</sub>O emissions (Phillips et al., 2001a). Ozone pollution decreased CH<sub>4</sub> emission (Morsky et al., 2008) while increase or decrease N<sub>2</sub>O 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 25 terrestrial ecosystems, it is essential to better understand the underlying mechanisms

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and attribute the variations in terrestrial ecosystem  $CH_4$  and  $N_2O$  fluxes to relative role

of various environmental factors.

#### 4.4 Uncertainties in this study and the way forward

We provided regional estimations of CH<sub>4</sub> and N<sub>2</sub>O 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. Firstly, only CH<sub>4</sub> production from DOC was considered in the current model. Other substrates may need to be included, for example, the CH<sub>4</sub> production from acetate could contribute nearly 20% to CH<sub>4</sub> production (Conrad, 1996; Mer and Roger, 2001). It might be better to include 10 more components in CH<sub>4</sub> 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 miss some possible high pulses in CH<sub>4</sub> and N<sub>2</sub>O fluxes at the time 15 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). Thirdly, 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 a research need for addi-
- tional 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<sub>4</sub> flux.

Fourthly, wetland area and classification could bring uncertainties to regional esti-

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mation of CH<sub>4</sub> and N<sub>2</sub>O fluxes. The fluxes of CH<sub>4</sub> and N<sub>2</sub>O 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. Fifthly, it is <sup>5</sup> 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<sub>2</sub>O emission from pasture may contribute a great proportion to the continental flux of N<sub>2</sub>O (Li et al., 1996). But N<sub>2</sub>O emission from man-10 aged pasture was not simulated in current model, due to a lack of spatially-explicit information on pasture management.

#### 5 Conclusions

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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 provided, to the best of our knowledge, the first continental-level simultaneous quantification and maps at 32 km×32 km resolution of annual CH<sub>4</sub> and N<sub>2</sub>O fluxes in North America's terrestrial ecosystems over the past three decades. While there are some uncertainties related to the estimation of CH<sub>4</sub> and N<sub>2</sub>O 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<sub>4</sub>

 $_{25}$  gas mitigation and management. To reduce uncertainties in regional estimation of CH<sub>4</sub> and N<sub>2</sub>O fluxes, it is greatly needed to further improve the representation of additional biogeochemical processes in the DLEM and the spatial data sets of wetland area and





pasture management. We also face the 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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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 <sub>CH40mMax</sub> (g C m <sup>-3</sup> day <sup>-1</sup> )	Aerobic	0.0207	0.0033-0.1306	(Segers, 1998)
1 TOTAL	Intermediate	0.4	0.0394-3.9418	(Segers, 1998)
$(a C m^{-3} dov^{-1})$	Anaerobic	0.75	0.0313-4.9624	(Segers, 1998) (Siteulo et al., 1005: Segrera, 1008: Sepri et al., 2004)
CH4OxidairMax		0.10	<0.001-103.7	(Sitaula et al., 1995, Segers, 1996, Saarret al., 2004)
$V_{\mathrm{CH}_{4_{\mathrm{OxidtransMax}}}}$ (g C m <sup>-3</sup> day <sup>-1</sup> )		0.5	0->51.84	(Segers, 1998)
V <sub>CH40</sub> (g C m <sup>-3</sup> day <sup>-1</sup> )		0.5	0->51.84	(Segers, 1998)
$Km_{CH_{4_{prod}}}^{-0.0130}$ (g C m <sup>-3</sup> )		10	1.68–91.2	(Lokshina et al., 2001)
$Km_{ m CH_{4_{Oxidair}}}$ (ppm)		10	5–18	(Nedwell and Watson, 1995; Arah and Stephen, 1998; Saari et al., 2004)
$Km_{_{CH_{4_{outdatans}}}}$ (g C m <sup>-3</sup> )		5	0.33–19.95	(Harrison, 1973;Joergensen, 1985; Linton and Buc- kee, 1977; Lamb and Garver, 1980; Nagai et al., 1973; O'Neill and Wilktnson, 1977; Ferenci et al., 1975)
$Km_{_{CH_{4_{outdatoll}}}}$ (g C m <sup>-3</sup> )		10	0.33–19.95	(Ferenci et al., 1975; Nagai et al., 1973; Linton and Buckee, 1977; Lamb and Garver, 1980; Joer- gensen, 1985; Harrison, 1973; O'Neill and Wilktn- son, 1977)
V <sub>max,denit</sub> (g N m <sup>-3</sup> day <sup>-1</sup> )	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*	
$K_{\text{densit}}$ (a N m <sup>-3</sup> )	Natural ecosystems	0.75	0.183–6.5	(Garcia-Ruiz et al., 1998: Yu et al., 2006)
(9.1.1.)				
	Cropland	1.5	1–10£	
$V_{\rm max,nit}$ (g N m <sup>-3</sup> day <sup>-1</sup> )	Natural ecosystems	0.02	0–2.18	(Kim et al., 1997; Sheibley et al., 2003)
	Cropland	0.05	0–5*	
$K_{\rm nit} (g \mathrm{N}\mathrm{m}^{-3})$	Natural ecosystems	0.75	0.21-1.11	(Sheibley et al., 2003)
	Cropland	1.5	1–10£	

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\* 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 other natural ecosystems.

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**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).

Vegetation type	Location	Reference
Tundra	68.63° N, 149.57° W	(Mosier et al., 1993)
Boreal broad leaf deciduous forest	62.85° N, 30.88° W	(Crill et al., 1994)
Boreal needle leaf evergreen forest	62.85° N, 30.88° W	(Crill et al., 1994)
Boreal needle leaf evergreen forest	53.92° N, 104.69° W	(Matson, 2008)
Temperate broad leaf deciduous forest	49,00° N, 8.00° E	(Koschorreck and Conrad, 1993)
Temperate broad leaf deciduous forest	49.17° N, 8.72° E	(Dörr et al., 1993)
Temperate broad leaf deciduous forest	46.6° N, 128.47° E	(Xiao et al., 2004)
Temperate broad leaf evergreen forest	46.6° N, 128.47° E	(Xiao et al., 2004)
Temperate needle leaf evergreen forest	29.0–30.33° N, 101.5–102.25° E	(Dong et al., 2003)
Tropical dry forest	1.00° S, 78.00° W	(Dörr et al., 1993)
Tropical rain forest	1.5° N, 18.0° E	(Tathy et al., 1992)
Temperate mixed forest	49,00° N, 8.00° E	(Koschorreck and Conrad, 1993)
Deciduous shrub	47.69° N, 133.52° E	(Song et al., 2009)
Evergreen shrub	40.95° N, 1.55° E	(Castaldi and Fierro, 2005)
C3 grassland	40.8° N, 104.75° W	(Mosier et al., 1996)
C4 grassland	40.83° N, 104.7° W	(Mosier et al., 2002)
Herbaceous wetland	47.69° N, 133.52° E	(Song et al., 2009)
Woody wetland	47.53° N, 93.47° W	(Dise, 1991)
Cropland	41.23° N, 103.00° W	(Kessavalou et al., 1998)
Desert	37° N, 116° W	(Strieg et al., 1992)

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**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).

Vegetation type	Location	Reference
Tundra	41.33° N, 106.33° W	(Sommerfeld et al., 1993)
Boreal broad leaf deciduous forest	40.02° N, 115.47° E	(Du et al., 2004)
Boreal needle leaf evergreen forest	53.92° N, 104.69° W	(Matson, 2008)
Temperate broad leaf deciduous forest	46.6° N, 128.47° E	(Xiao et al., 2004)
Temperate broad leaf evergreen forest	46.6° N, 128.47° E	(Xiao et al., 2004)
Temperate needle leaf evergreen forest	29.0~30.33° N, 101.5~102.25° E	(Dong et al., 2003)
Tropical dry forest	10.5° S, 62.5° W	(Garcia-Montiel et al., 2002)
Tropical rain forest	10.5° S, 62.5° W	(Garcia-Montiel et al., 2002)
Temperate mixed forest	46.6° N, 128.47° E	(Xiao et al., 2004)
Deciduous shrub	47.69° N, 133.52° E	(Song et al., 2009)
Evergreen shrub	46.37°–46.56° N, 119.47°~119.78° W	(Mummey et al., 1997)
C3 grassland	43.03° N, 119.15° E	(Huang et al., 2003)
C4 grassland	40.83° N, 104.7° W	(Mosier et al., 2002)
Herbaceous wetland	47.69° N, 133.52° E	(Song et al., 2009)
Woody wetland	53.63° N, 106.20° W	(Matson, 2008)
Cropland	45.67° N, 111.15° W	(Dusenbury et al., 2008)
Cropland	41.23° N, 103.00° W	(Kessavalou et al., 1998)
Desert	36.82° N,115.92° W	(Billings et al., 2002)

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

Major	V <sub>CH4proMax</sub>	V <sub>CH4OvidaidMax</sub>	V <sub>CH4000ttrane</sub>	V <sub>CH4Ovideoilmax</sub>	Km <sub>CH4nrod</sub>	Km <sub>CH4Ovidair</sub>	Km <sub>CH4nvidtrane</sub>	Km <sub>CH4-minteroil</sub>
ecosystem type	(g C m <sup>-3</sup> day <sup>-1</sup> )	(g C m <sup>-3</sup> )	(ppm)	(g C m <sup>-3</sup> )	(g C m <sup>-3</sup> )			
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

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

Major ecosystem type	V <sub>denimax</sub> (g Nm <sup>-3</sup> day <sup>-1</sup> )	K <sub>deni</sub> (g Nm <sup>-3</sup> )	V <sub>nitmax</sub> (g Nm <sup>-3</sup> day <sup>-1</sup> )	K <sub>nit</sub> (g Nm <sup>−3</sup> )
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

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**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 (TgCa <sup>-1</sup> )	-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	$0.19 \pm 0.01$	$0.56 \pm 0.07$	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.

Method	Period	Domain	$CH_4 (T g C a^{-1})$	N <sub>2</sub> O (T g N a <sup>-1</sup> )	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.

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**Table 8.** Comparison of the DLEM-estimated  $CH_4$  emission rate (g C m<sup>-2</sup> a<sup>-1</sup>) 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		-0.0550.575	-0.109	-0.112	-0.075
Boreal forest	-0.128±0.010		-0.0740.430	-0.125	-0.198	-0.140
Temperate forest	-0.178±0.012		-0.068-01.15	-0.155	-0.428	-0.181
Tropical dry forest	-0.244±0.013		-0.0820.520	-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 floodblains			
Woody wetland	$7.871 \pm 0.807$		·····			

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**Table 9.** Comparison of the DLEM-estimated N<sub>2</sub>O emission rate  $(gNm^{-2}a^{-1})$  with other studies at biome level (Positive values represent N<sub>2</sub>O emission).

Biome type	This study	(Potter et al., 1996)	Recalculated from Xu et al. (2008)
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	$0.220 \pm 0.030$	0.081**	0.010-0.725
Herbaceous wetland	0.169±0.014		0.002-0.251
Woody wetland	$0.053 \pm 0.005$		0.002–0.251

\* Temperate grassland and tropical savanna and grassland; \*\* without fertilization.



**Fig. 1.** Conceptual model of the Dynamic Land Ecosystem Model (DLEM) (five core components were included in the DLEM).

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Major processes: Aoxid: Atmospheric CH<sub>4</sub> oxidation; CH4pro: CH<sub>4</sub> production; CH4oxid: CH<sub>4</sub> Oxidation during diffusion and ebullition transport; CH4oxidp: CH4 oxidation during plantmediated transport (Occur in herbaceous wetland only); Dif: CH<sub>4</sub> diffusion transport; Ebu: CH<sub>4</sub> ebullition transport; Pmt: Plant-mediated transport of CH<sub>4</sub>; (Occur in herbaceous wetland only); Nitri: Nitrification; Denitri: Denitrification;

DLEM provides the environment factors and substrate for  $\rm CH_4$  and  $\rm N_2O$  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 denitrification are considered in the  $N_2O$  module).



Fig. 3. Contemporary vegetation map used in this study (the year of 2000 was shown).

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**Fig. 4.** Comparison of the DLEM-estimated CH<sub>4</sub> and N<sub>2</sub>O fluxes with field observations (**A**) CH<sub>4</sub> flux in Durham Forest (42° N, 73° W) (TRAGNET); (**B**) CH<sub>4</sub> flux in Alaska wetland (64.8° N, 147.7° W) (TRAGNET); (**C**) CH<sub>4</sub> flux in Sallie fen (43.21° N, 71.05° W); (**D**) CH<sub>4</sub> flux in Hubbard Brook Forest (43.95° N, 71.74° W) (Groffman et al., 2006, 2009); (**E**) N<sub>2</sub>O flux in Hubbard Brook Forest (43.95° N, 71.74° W) (Groffman et al., 2006, 2009); (**E**) N<sub>2</sub>O 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<sup>-2</sup> day<sup>-1</sup> 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).

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**Fig. 5.** Temporal patterns of  $CH_4$  and  $N_2O$  fluxes in North America's terrestrial ecosystems during 1979–2008.



Fig. 6. Spatial distribution of (A)  $CH_4$  and (B)  $N_2O$  fluxes in North America's terrestrial ecosystems during 1979–2008.



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**Fig. 8.** Comparison of the DLEM-derived N<sub>2</sub>O fluxes with the estimations by an empirical model (Xu et al., 2008). Spatial correlation between the 21-year average of these two dataset shows a high correlation coefficient of 0.54 (the regression model is: the DLEM-derived N<sub>2</sub>O = 0.8887 \* Empirically-modeled N<sub>2</sub>O,  $R^2$  = 0.39; P < 0.01).











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