



# Multifactor controls on terrestrial N<sub>2</sub>O flux over North America from 1979 through 2010

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**Abstract.** Nitrous oxide (N<sub>2</sub>O) is a potent greenhouse gas which also contributes to the depletion of stratospheric ozone (O<sub>3</sub>). However, the magnitude and underlying mechanisms for the spatiotemporal variations in the terrestrial sources of N<sub>2</sub>O are still far from certain. Using a process-based ecosystem model (DLEM – the Dynamic Land Ecosystem Model) driven by multiple global change factors, including climate variability, nitrogen (N) deposition, rising atmospheric carbon dioxide (CO<sub>2</sub>), tropospheric O<sub>3</sub> pollution, N fertilizer application, and land conversion, this study examined the spatial and temporal variations in terrestrial N<sub>2</sub>O flux over North America and further attributed these variations to various driving factors. From 1979 to 2010, the North America cumulatively emitted  $53.9 \pm 0.9$  Tg N<sub>2</sub>O-N ( $1 \text{ Tg} = 10^{12} \text{ g}$ ), of which global change factors contributed  $2.4 \pm 0.9$  Tg N<sub>2</sub>O-N, and baseline emission contributed  $51.5 \pm 0.6$  Tg N<sub>2</sub>O-N. Climate variability, N deposition, O<sub>3</sub> pollution, N fertilizer application, and land conversion increased N<sub>2</sub>O emission while the elevated atmospheric CO<sub>2</sub> posed opposite effect at continental level; the interactive effect among multiple factors enhanced N<sub>2</sub>O emission over the past 32 yr. N input, including N fertilizer application in cropland and N deposition, and multi-factor interaction dominated the increases in N<sub>2</sub>O emission at continental level. At country level, N fertilizer application and multi-factor interaction made large contribution to N<sub>2</sub>O emission increase in the United States of America (USA). The climate variability dominated the increase in N<sub>2</sub>O emission from Canada. N inputs and multiple factors interaction made large contribution to the increases in N<sub>2</sub>O emission from Mexico. Central and southeastern parts of the North America – including

central Canada, central USA, southeastern USA, and all of Mexico – experienced increases in N<sub>2</sub>O emission from 1979 to 2010. The fact that climate variability and multi-factor interaction largely controlled the inter-annual variations in terrestrial N<sub>2</sub>O emission at both continental and country levels indicate that projected changes in the global climate system may substantially alter the regime of N<sub>2</sub>O emission from terrestrial ecosystems during the 21st century. Our study also showed that the interactive effect among global change factors may significantly affect N<sub>2</sub>O flux, and more field experiments involving multiple factors are urgently needed.

## 1 Introduction

Nitrous oxide (N<sub>2</sub>O) plays an important role in both contributing to the greenhouse effect (Denman et al., 2007; Rodhe, 1990) and depleting stratospheric ozone (O<sub>3</sub>) (Denman et al., 2007; Cicerone, 1987). The atmospheric N<sub>2</sub>O concentration has increased from 270 ppb (one part per billion) in 1750 to 319 ppb in 2005 (Forster et al., 2007); terrestrial ecosystems under the impacts of anthropogenic activities have been recognized as one of major sources for this increase (Keller et al., 1986; Bouwman et al., 1993; Del Grosso et al., 2006; Li et al., 1996; Liu, 1996; Repo et al., 2009; Williams et al., 1992; Forster et al., 2007; Denman et al., 2007; Song et al., 2009). Quantifying the magnitude of terrestrial N<sub>2</sub>O flux and the underlying mechanisms will be crucial for advancing our understanding on the dynamics of atmospheric N<sub>2</sub>O concentration, and further providing helpful information for policy-makers to curb the

continuous increase in atmospheric N<sub>2</sub>O concentration (Tian et al., 2010b; Denman et al., 2007).

N<sub>2</sub>O flux has been recognized as a result of a suite of microbial processes influenced by a variety of environmental factors (Conrad, 1996; Williams et al., 1992; Pilegaard et al., 2006). Global change will alter these environmental factors and substrates, and further change the N<sub>2</sub>O flux (Bouwman et al., 1993; Conrad, 1996; Goldberg and Gebauer, 2009; Kanerva et al., 2007; Kettunen et al., 2005; Williams et al., 1992; Ambus and Robertson, 1999). For example, nitrogen (N) input may stimulate N<sub>2</sub>O production by increasing substrate availability (Kettunen et al., 2005; Mcswiney and Robertson, 2005); elevated atmospheric CO<sub>2</sub> may reduce N availability in soil owing to progressive N accumulation in plant biomass (Luo et al., 2004; McGuire et al., 1995), which inhibit the N<sub>2</sub>O emission (Phillips et al., 2001); alternatively, elevated atmospheric CO<sub>2</sub> might increase photosynthetic products and stimulate microbial process, and thus increase N<sub>2</sub>O emission (Kettunen et al., 2005; Ineson et al., 1998). If these two effects are counterbalanced, it may appear as neutral response of N<sub>2</sub>O flux to elevated atmospheric CO<sub>2</sub> (Kanerva et al., 2007; Ambus and Robertson, 1999). Tropospheric O<sub>3</sub> pollution may alter microbial community (Kanerva et al., 2008) and cause an increase or decrease in N<sub>2</sub>O emission, depending on time and location (Kanerva et al., 2007). By comparison, the effects of climate variability and land conversion on the N<sub>2</sub>O emission are more complicated, largely replying upon the specific site condition (Jiang et al., 2009; Goldberg and Gebauer, 2009; Zhang et al., 2007b).

In the past decades, considerable emphasis has been put on the accurate estimation of terrestrial N<sub>2</sub>O flux (Potter et al., 1996; Xu et al., 2008; Liu, 1996; Denman et al., 2007; Matson and Vitousek, 1990; Bouwman et al., 1993). A number of estimates for N<sub>2</sub>O flux have been achieved by extrapolating average fluxes from chamber-based measurements to the areal extent of vegetation or soil classes from which the measurements were taken (Keller et al., 1986; Huang et al., 2003; Matson et al., 1989), or by using a simple empirical model (Xu et al., 2008). In these approaches, uncertainties were introduced because the spatial heterogeneity within the classes, as well as the seasonal and inter-annual variabilities in climatic and biotic controls on emission rates, were, at least partially neglected (Potter et al., 1996; Matson et al., 1989). Meanwhile, the empirical methods in estimating regional N<sub>2</sub>O flux could not be used to attribute the spatiotemporal variations in terrestrial N<sub>2</sub>O flux to environmental drivers. Therefore, a large-scale estimation of terrestrial N<sub>2</sub>O flux with consideration of spatial heterogeneity of soil, vegetation, and climate variations that could be used for factorial attribution is highly needed.

The process-based modeling approach is gaining popularity in estimating regional N<sub>2</sub>O flux (Del Grosso et al., 2006; Li et al., 2001; Potter et al., 1996). Although process-based models have the potential to explore the relative contributions

of each driving force to the spatiotemporal variations in terrestrial N<sub>2</sub>O, as they have been used for terrestrial CO<sub>2</sub> flux (McGuire et al., 2001; Tian et al., 2003; Mu et al., 2008) and CH<sub>4</sub> flux (Xu et al., 2010), none of them have been utilized to attribute the spatial and temporal variations in terrestrial N<sub>2</sub>O flux to its driving factors.

North America, one of the extensively investigated continents, still lacks accurate estimates due to limitations in methodology and observations (Kort et al., 2008). A number of studies estimated the N<sub>2</sub>O emission by solely focusing on one ecosystem type or considering one global change factor (Del Grosso et al., 2006; Xu et al., 2008; Potter et al., 1996). For example, Li et al. (1996) estimated N<sub>2</sub>O emission from cropland in the United State of America (USA) by using a process-based model DNDC (DeNitrification DeComposition: Li et al., 1996). Del Grosso et al. (2006) estimated the N<sub>2</sub>O flux from cropland in the USA by using the DAYCENT model (Daily Century Model: Del Grosso et al., 2006). One of our previous studies simulated the regional flux of N<sub>2</sub>O over North America during 1979–2008 by using a process-based ecosystem model DLEM, driven by multiple global change factors including climate variability, elevated atmospheric CO<sub>2</sub>, N deposition, O<sub>3</sub> pollution, N fertilizer application, and land use change (Tian et al., 2010b). In this study, we will extend our previous work and attribute the spatial and temporal variations in terrestrial N<sub>2</sub>O flux to various driving factors.

Specifically, the objectives of this study are: (1) to examine the factorial contributions to the spatial variation of terrestrial N<sub>2</sub>O flux over North America during 1979–2010; (2) to quantify the factorial contributions to the temporal variations in terrestrial N<sub>2</sub>O flux over North America during 1979–2010; and (3) to quantify the factorial contributions to the 32-yr cumulative flux of N<sub>2</sub>O over North America at both continental and country levels. The global change factors evaluated in this study include climate variability, rising atmospheric CO<sub>2</sub>, N deposition, O<sub>3</sub> pollution, changes in land use and land cover type, and N fertilizer application in cropland. The interactive effects among these six factors were calculated as the difference between simulated changes in N<sub>2</sub>O flux driven by all factors together and changes in N<sub>2</sub>O flux caused by six individual factors (see Experiment design section for the detail information).

## 2 Materials and methods

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

The model used in this study is a process-oriented ecosystem model DLEM, which couples major biogeochemical cycles, hydrological cycles, and vegetation dynamics to make daily, spatially-explicit estimates of carbon (C), N, and water fluxes and pool sizes in terrestrial ecosystems (Tian et al., 2010b). The DLEM also simulates the managed ecosystems,

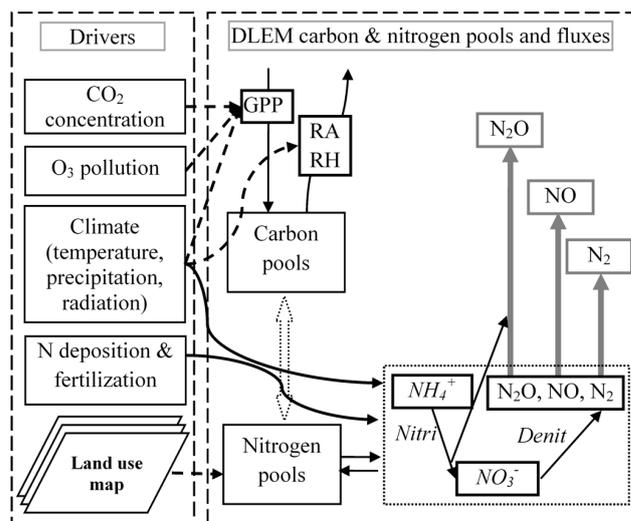
including agricultural ecosystems, plantation forests, and pastures. The spatial data set of land management practices, such as irrigation, fertilization, rotation, and harvest, can be used as input information for simulating influences of land management on the structure and functioning of ecosystems. This model has been calibrated against various field data from the Chinese Ecological Research Network (CERN), US Long-Term Ecological Research (LTER) network, and AmeriFlux network, which cover various ecosystems, including forest, grassland, shrubland, tundra, desert, natural wetlands, and cropland. The simulated results have been compared with independent field data and satellite products. The DLEM operates at a daily time step and at varied spatial resolutions, from meters to kilometers, from regional to global. The detailed information for DLEM has been described in our previous publications (Liu et al., 2008; Ren et al., 2007, 2011a, b; Zhang et al., 2007a; Tian et al., 2010a, b, 2011a, b; Xu et al., 2010; Lu et al., 2012), and the N<sub>2</sub>O module has been described in detail in Tian et al. (2010b).

In the DLEM, the N<sub>2</sub>O module is incorporated into nitrogen cycling; it simulates the nitrification and denitrification processes. Both denitrification and nitrification processes are simulated as one-step process as we do not consider the mid-products in each process. Nitrification, a process converting ammonium into nitrate, is simulated as a function of soil temperature, moisture, and the NH<sub>4</sub><sup>+</sup> concentration (Lin et al., 2000; Tian et al., 2010b). It should be noted that a small portion of N<sub>2</sub>O is from nitrification processes. Denitrification, through which the nitrate is converted into N-containing gases, is simulated in the DLEM as a function of soil temperature, moisture, and the NO<sub>3</sub><sup>-</sup> concentration (Lin et al., 2000). The empirical equation reported by Davidson et al. (2000) is used to separate N<sub>2</sub>O from other gases (mainly NO and N<sub>2</sub>).

In summary, multiple global change factors directly and/or indirectly affect N<sub>2</sub>O processes in the DLEM (Fig. 1). The elevated atmospheric CO<sub>2</sub> and O<sub>3</sub> pollution yield indirect impacts on N<sub>2</sub>O fluxes through their effects on carbon processes and carbon-nitrogen interaction; the climate variability yield direct and/or indirect impacts on N<sub>2</sub>O flux; N deposition and N fertilizer application directly affect N<sub>2</sub>O flux since they will change available N in soil for N<sub>2</sub>O production, and indirectly impact carbon process and C-N interaction; the effect of land conversion is complicated since it might change all the ecosystem properties and hence N<sub>2</sub>O flux. It should be noted that there are other environmental factors, for example, soil pH, and soil porosity, etc., that might influence N<sub>2</sub>O flux.

## 2.2 Study area and input data

This study mainly focused on North America, which includes the USA, Canada, and Mexico, covering a total area of approximately 24.71 million km<sup>2</sup>, approximately 4.8 % of the planet's surface or 16.5 % of its land area. Excluding water bodies, North America consists of 21 237 grids at a spatial



Major processes: *Nitri*: Nitrification; *Denit*: Denitrification; GPP is the gross primary productivity; RA is the autotrophic respiration from plant, and RH is the heterotrophic respiration; Drivers are the multiple global change factors which yield controls on or feedback to ecosystem processes in the DLEM framework. The effects from drivers were expressed as the line starting from drivers to ecosystem processes or pools. Solid lines represent direct, while dash lines represent indirect impacts on N<sub>2</sub>O processes.

**Fig. 1.** Conceptual diagram showing major processes for N<sub>2</sub>O flux in response to multiple global change factors in the DLEM model (Only nitrification and denitrification processes are shown in the figure; other nitrogen processes including nitrogen fixation, mineralization, immobilization etc. are not shown in this figure since they are not the focus of this study; see Tian et al. (2010b) for detailed information).

resolution of 32 km by 32 km, which is consistent with the North American Regional Reanalysis (NARR) dataset.

We developed gridded (32 × 32 km), geo-referenced, time-series data sets of climate (including daily average, maximum, and minimum temperatures, precipitation, humidity, and solar radiation), annual N deposition rate, annual land-cover change and land management practices (including fertilization, irrigation) for the entire North America. The data development has been described in detail in a previous publication (Xu et al., 2010). The same dataset was extended to cover the time period of 1900–2010 and used in this study. The climate data was extended to 2010 by processing the NARR dataset (Mesinger et al., 2006). The land use and land cover change data, N fertilizer data, O<sub>3</sub> pollution data, and N deposition were assumed unchanged after 2005; the N fertilizer data and N deposition were assumed unchanged after 2008. The annual atmospheric concentration of CO<sub>2</sub> was updated based on the dataset from National Oceanic and Atmospheric Administration (NOAA) ([www.esrl.noaa.gov](http://www.esrl.noaa.gov)).

Historical data from 1901 to 2010 are used to drive the transient model simulations in this study. The transient input

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

Simulation	Climate	Nitrogen deposition	CO <sub>2</sub>	O <sub>3</sub>	Nitrogen fertilizer	Land conversion
1	1900–2010	1900–2010	1900–2010	1900–2010	1900–2010	1900–2010
2	1900–1979	1900–1979	1900–1979	1900–1979	1900–1979	1900–1979
3	1900–2010	1900	1900	1900	1900	1900
4	1900–1979	1900	1900	1900	1900	1900
5	1900	1900–2010	1900	1900	1900	1900
6	1900	1900–1979	1900	1900	1900	1900
7	1900	1900	1900–2010	1900	1900	1900
8	1900	1900	1900–1979	1900	1900	1900
9	1900	1900	1900	1900–2010	1900	1900
10	1900	1900	1900	1900–1979	1900	1900
11	1900	1900	1900	1900	1900–2010	1900
12	1900	1900	1900	1900	1900–1979	1900
13	1900	1900	1900	1900	1900	1900–2010
14	1900	1900	1900	1900	1900	1900–1979

Note: 1900–2010 indicates that the data for the time period of 1900–2010 was used in the simulation; while 1900–1979 indicates that the data for the time period of 1900–1979 was used in the simulations and the simulations after 1979 was fed by the data of 1979.

data include: (1) daily climate data from 1901 to 2010, including maximum, minimum and average temperatures, relative humidity, solar radiation, and precipitation; the data from 1901 to 1978 were randomly assigned as one year during 1979–2010; (2) annual N deposition from 1901 to 2010; (3) annual O<sub>3</sub> pollution data from 1901 to 2010; (4) atmospheric CO<sub>2</sub> concentration from 1901 to 2010; (5) cropland and urban distribution from 1901 to 2005 – the land use since 2005 was assumed unchanged due to shortage of data; and (6) N fertilizer application data for cropland over the time period of 1901–2010.

### 2.3 Experimental design

In this study, we performed fourteen simulation experiments to determine the relative effects of N deposition, O<sub>3</sub> pollution, climate variability, elevated atmospheric CO<sub>2</sub>, land-use change, and N fertilizer application on terrestrial N<sub>2</sub>O flux over North America. One overall simulation was set up to simulate the terrestrial N<sub>2</sub>O flux over North America by considering the temporal and spatial dynamics of all six global change factors. Six additional simulations were set up to simulate the effects of each individual factor on N<sub>2</sub>O flux. For example, to determine the effects of climate variability alone, we ran DLEM using the gridded historical daily data for air temperature, including maximum, minimum, and average air temperatures, relative humidity, solar radiation, and precipitation; but kept all other five global change factors at the levels of 1900: the atmospheric CO<sub>2</sub> concentration, N deposition, O<sub>3</sub> pollution, N fertilizer application for cropland, and the land cover type (potential vegetation map with cropland and urban land in 1900). To determine the effects of CO<sub>2</sub> fertilization alone, we ran DLEM using the historical atmospheric CO<sub>2</sub> concentrations, but kept all other five global change factors constant: a 30-yr averaged daily climate data

was used to represent the mean climate condition, the potential vegetation map with crop and urban land in 1900 was used to represent the constant land cover type, and the N deposition, O<sub>3</sub> pollution, and N fertilizer application data were kept constant in the year of 1900. For each of the above seven simulations, we set up one corresponding simulation except the input data in 1979 was used to drive the post-1979 simulations. This simulation design was used to capture the internal dynamics of the system, which served as baseline; the difference between each pair of simulation is defined as the factor-induced N<sub>2</sub>O flux. For example, the time-series difference between simulation #1 and simulation #2 for 1979–2010 is caused by all global change factors and their interaction because the internal system dynamic defined as results from simulation #2 was excluded (Table 1).

The implementation of DLEM simulation included the following steps: (1) equilibrium run, (2) spinning-up run, and (3) transient run. In this study, we used potential vegetation, long-term mean climate during 1979–2010, the levels of N deposition, O<sub>3</sub>, and atmospheric CO<sub>2</sub> concentration in the year 1900 to drive the model simulations to an equilibrium state (i.e. the inter-annual variations are <0.1 g C m<sup>-2</sup> for carbon storage, <0.1 g N m<sup>-2</sup> for N storage). After the system reached an equilibrium state, the model was run with an addition of cropland and urban areas for another 3000 yr for spinning-up purposes. Finally, the model was run in transient mode with daily climate data, annual CO<sub>2</sub> concentration, and N deposition inputs from 1901 to 2010 to simulate the terrestrial N<sub>2</sub>O flux. The outputs between 1979 and 2010 were analyzed to show the spatial and temporal patterns of terrestrial N<sub>2</sub>O flux in North America's terrestrial ecosystems. Urban was treated as grassland, which is the same strategy used in other terrestrial biosphere models. Baseline flux was defined as the terrestrial N<sub>2</sub>O flux during 1979–2010

**Table 2.** Changing rates of driving factors from 1979 to 2010 for DLEM simulations.

Variables	Changing rates (Mean $\pm$ SD)	
Climate	Maximum temperature ( $^{\circ}\text{C a}^{-1}$ )	$0.04 \pm 0.01^*$
	Minimum temperature ( $^{\circ}\text{C a}^{-1}$ )	$0.03 \pm 0.01^*$
	Average temperature ( $^{\circ}\text{C a}^{-1}$ )	$0.03 \pm 0.01^*$
	Precipitation ( $\text{mm a}^{-1}$ )	$0.09 \pm 0.62$
	Relative humidity ( $\% \text{ a}^{-1}$ )	$-0.01 \pm 0.01$
	Solar radiation ( $\text{W m}^{-2} \text{ a}^{-1}$ )	$0.19 \pm 0.03^*$
Others	Tropospheric O <sub>3</sub> pollution ( $\text{ppm-h a}^{-1}$ )	$0.96 \pm 0.08^*$
	N deposition ( $\text{mg m}^{-2} \text{ a}^{-1}$ )	$1.96 \pm 0.05^*$
	N fertilizer application ( $\text{mg m}^{-2} \text{ a}^{-1}$ )	$0.02 \pm 0.01^*$
	Atmospheric CO <sub>2</sub> concentration ( $\text{ppm a}^{-1}$ )	$1.68 \pm 0.02^*$

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

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

Plant functional type	Tundra	Forest	Shrub	Grassland	Wetland	Desert and others	Cropland
Area (million km <sup>2</sup> )	4.05	6.93 ~ 6.99	3.57 ~ 3.59	2.61 ~ 2.64	2.06 ~ 2.07	0.53 ~ 0.60	2.51 ~ 2.59
Percentage	18.09	31.10	15.98	11.72	9.23	2.49	11.39

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

simulated by the DLEM driven by the input data of 1979. The changes thereafter compared to baseline flux were assumed solely caused by global change factors, individually or in combination.

## 2.4 Model parameterization

The model parameterization and validation at both site and regional levels were documented in our previous publication (Tian et al., 2010b). We will not describe them in detail in this paper.

## 2.5 Statistic method

The regression analysis was used in this study to quantify the long-term changing trend of input data and terrestrial N<sub>2</sub>O fluxes estimated by various simulations. All the statistical analyses were conducted using the R program 12.0 for Windows XP.

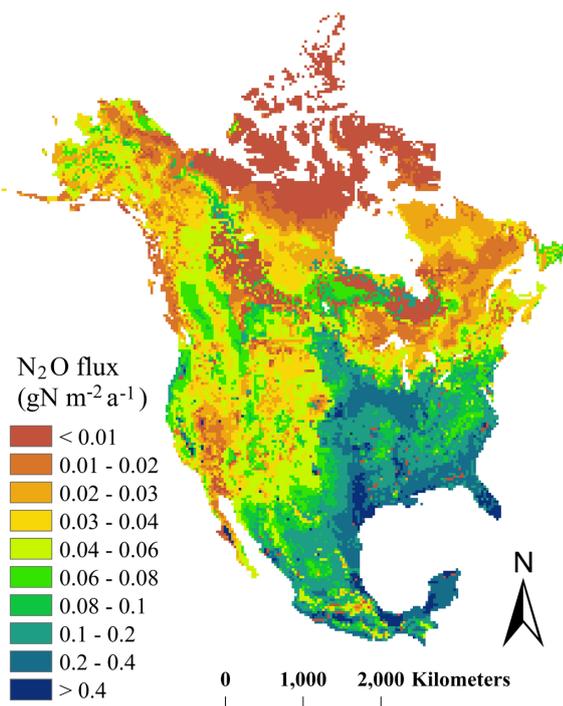
## 3 Results

### 3.1 Environmental changes over North America during 1979–2010

For the climate variables, maximum, minimum, and average temperatures, and solar radiation showed significantly increasing trends at the rates of  $0.04 \pm 0.01$   $^{\circ}\text{C a}^{-1}$ ,  $0.03 \pm 0.01$   $^{\circ}\text{C a}^{-1}$ ,  $0.03 \pm 0.01$   $^{\circ}\text{C a}^{-1}$ , and  $0.19 \pm$

$0.03 \text{ W m}^{-2} \text{ a}^{-1}$ , respectively; yet precipitation and relative humidity did not show any significantly changing trends over the study period. All the other driving factors significantly increased since 1979; the long-term increasing rates were  $0.96 \pm 0.08$   $\text{ppm-h a}^{-1}$  for tropospheric O<sub>3</sub> pollution,  $1.96 \pm 0.05$   $\text{mg m}^{-2} \text{ a}^{-1}$  for N deposition,  $0.02 \pm 0.01$   $\text{g m}^{-2} \text{ a}^{-1}$  for N fertilizer application, and  $1.68 \pm 0.02$   $\text{ppm a}^{-1}$  for atmospheric CO<sub>2</sub> concentration, respectively (Table 2). The areas of different land cover types changed slightly throughout the study period; the cropland area increased from 2.51 million km<sup>2</sup> to 2.59 million km<sup>2</sup>; the area of forest, shrubland, grassland and wetland changed in a very small magnitude (Table 3).

Spatial variations of input data were shown in a previous paper (Xu et al., 2010). Normally, the severely O<sub>3</sub>-polluted area over North America locates in western part of North America such as the northwestern USA which could be as high as more than 5000 ppb h<sup>-1</sup> (monthly cumulated hourly O<sub>3</sub> dose over a threshold of 40 ppb in ppb-h), while the other areas, especially northern end of continental North America, were featured by low tropospheric O<sub>3</sub> pollution. The regions with high N fertilizer application (larger than  $10 \text{ g N m}^{-2} \text{ a}^{-1}$ ) concentrate in USA, including western, central, and eastern coastal areas of USA. The Canada and Mexico had a small amount of cropland and received lower application rate of N fertilizer. The high N deposition primarily occurred in eastern part of the continental North America, including southeastern Canada, eastern USA and portions of Mexico (larger than  $1 \text{ g N m}^{-2} \text{ a}^{-1}$ ), while the northern



**Fig. 2.** Spatial variations of terrestrial N<sub>2</sub>O fluxes caused by global change factors over North America from 1979 to 2010.

Canada was featured by quite low N deposition (lower than  $0.01 \text{ g N m}^{-2} \text{ a}^{-1}$ ).

### 3.2 Spatial distribution of N<sub>2</sub>O flux over North America during 1979–2010

The terrestrial N<sub>2</sub>O flux over North America showed a significant spatial variation, with a strong source in southeastern continental North America, including southeastern USA and the majority of Mexico, and a weak source in the northern part of North America (Fig. 2). At the country level, central and southeastern USA featured high N<sub>2</sub>O emission, up to nearly  $1 \text{ g N m}^{-2} \text{ a}^{-1}$ , while western USA had relatively low N<sub>2</sub>O emission rates. There was a north-to-south increasing gradient of N<sub>2</sub>O emission across Canada: the N<sub>2</sub>O emission rate was as low as  $0.0001 \text{ g N m}^{-2} \text{ a}^{-1}$  in northern Canada, and as high as nearly  $0.4 \text{ g N m}^{-2} \text{ a}^{-1}$  in southern Canada; the entire Mexico acted as a strong source for N<sub>2</sub>O, with the national N<sub>2</sub>O emission being  $>0.1 \text{ g N m}^{-2} \text{ a}^{-1}$  over the past 32 yr.

### 3.3 Factorial contribution to the spatial variations in terrestrial N<sub>2</sub>O flux during 1979–2010

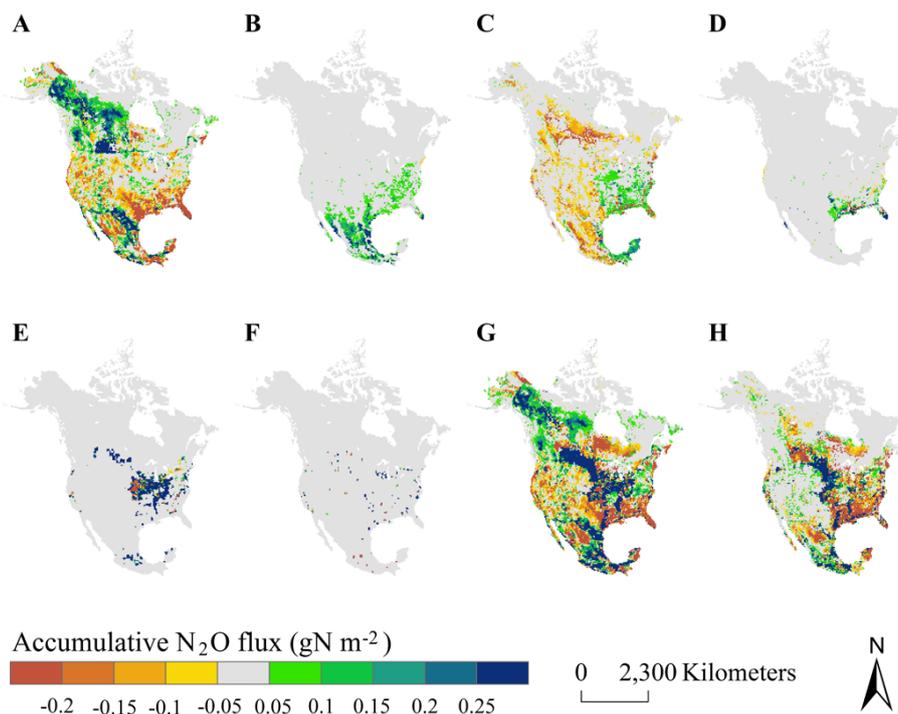
Nitrogen release as N<sub>2</sub>O is one of major pathways for nitrogen loss from terrestrial ecosystem. Throughout the study period, the cumulative N<sub>2</sub>O emission over North America was composed of two components: one is the background emission, defined as the N<sub>2</sub>O flux during 1979–2010 sim-

ulated by the DLEM driven by the input data of 1979; the other is the flux contributed by changes in various individual and interactive effects of several global change factors. After removing the baseline flux of N<sub>2</sub>O, the remaining changes in N<sub>2</sub>O fluxes over the study period are resulted from six global change factors and their interaction.

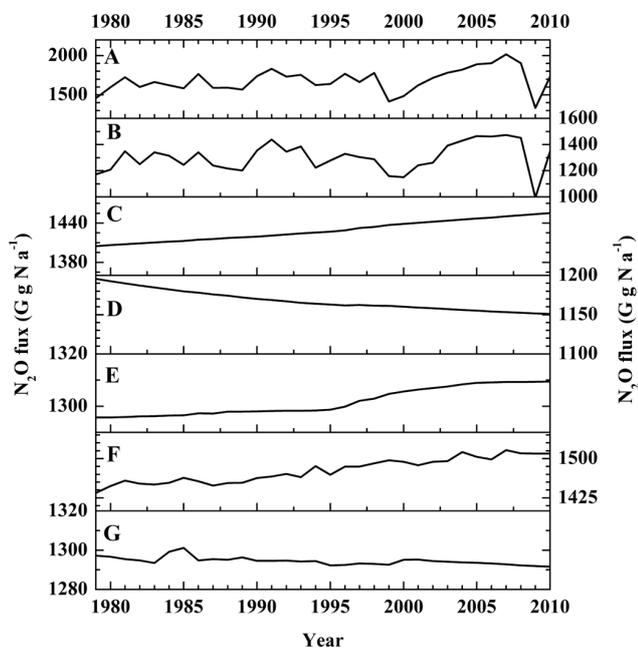
Over the 32-yr study period, climate variability enhanced N<sub>2</sub>O emission over the majority of central Canada, while decreased N<sub>2</sub>O emissions over portions of western Alaska, majority of the USA, and a portion of Mexico (Fig. 3a). N deposition enhanced N<sub>2</sub>O emission over the entire continental North America, with prominent increases over southeastern USA and Mexico (Fig. 3b). The elevated atmospheric CO<sub>2</sub> enhanced N<sub>2</sub>O emissions in southeastern USA and a portion of Mexico, while decreased N<sub>2</sub>O emissions in western USA, central Canada, and the majority of Mexico (Fig. 3c). The O<sub>3</sub> pollution increased N<sub>2</sub>O emissions in portions of southeastern USA, while no significant impacts on N<sub>2</sub>O flux were found in other areas (Fig. 3d). The N fertilizer application enhanced N<sub>2</sub>O emission in central USA, which is agricultural land (Fig. 3e). Land conversion only affected a small amount of area that experienced land use conversion between natural vegetation and cropland or urban in the past few years (Fig. 3f). The interactive effect among global change factors enhanced N<sub>2</sub>O emissions in central USA, while decreased N<sub>2</sub>O emissions in southeastern USA and central Canada (Fig. 3h). Combining all the effects from various global change factors, the N<sub>2</sub>O emissions were enhanced across central Canada and central USA and portions of Mexico, yet were decreased in western and southeastern USA and eastern Canada (Fig. 3g).

### 3.4 Temporal patterns of N<sub>2</sub>O flux over North America during 1979–2010

The continental-level terrestrial flux of N<sub>2</sub>O over North America showed a significant inter-annual fluctuation during 1979–2010 (Fig. 4). The mean annual N<sub>2</sub>O flux over North America's terrestrial ecosystems was  $1.68 \pm 0.15 \text{ Tg N}_2\text{O-N a}^{-1}$ , with an overall increasing rate of  $5.47 \pm 2.74 \text{ Gg N}_2\text{O-N a}^{-1}$  ( $1 \text{ Gg} = 10^9 \text{ g}$ ) over 32 yr ( $P = 0.055$ ). The long-term increasing trend and inter-annual fluctuation in terrestrial N<sub>2</sub>O flux resulted from multiple global change factors (Fig. 4). Climate variability exerted a significant inter-annual variation in N<sub>2</sub>O flux, with a slightly increasing rate of  $2.72 \pm 2.07 \text{ Gg N}_2\text{O-N a}^{-1}$  ( $P = 0.199$ ). N deposition contributed to an increasing rate of  $1.67 \pm 0.03 \text{ Gg N}_2\text{O-N a}^{-1}$  ( $P < 0.001$ ), and N fertilizer application contributed to an increasing rate at  $2.39 \pm 0.13 \text{ Gg N}_2\text{O-N a}^{-1}$  for continental N<sub>2</sub>O emission ( $P < 0.001$ ). O<sub>3</sub> pollution contributed to an increasing rate of  $0.54 \pm 0.03 \text{ Gg N}_2\text{O-N a}^{-1}$  ( $P < 0.001$ ). The increased atmospheric CO<sub>2</sub> concentration continuously decreased N<sub>2</sub>O emission, which generated a decreasing rate of  $1.33 \pm 0.06 \text{ Gg N}_2\text{O-N a}^{-1}$  ( $P < 0.001$ ) over the study



**Fig. 3.** Factorial contributions to the spatial variations in cumulated N<sub>2</sub>O flux over North America from 1979 to 2010 (A: climatic variability; B: N deposition; C: CO<sub>2</sub>; D: tropospheric O<sub>3</sub> pollution; E: N fertilizer application; F: land conversion; G: all combined; H: interaction).



**Fig. 4.** Temporal variations of terrestrial N<sub>2</sub>O flux caused by global change factors over North America from 1979 to 2010 (A: all combined simulation; B: climate only simulation; C: N deposition only simulation; D: CO<sub>2</sub> only simulation; E: tropospheric O<sub>3</sub> pollution only simulation; F: N fertilizer application only simulation; G: land conversion only).

period, while land conversion did not yield significant changing trends of N<sub>2</sub>O emission over the study period.

### 3.5 Factorial contributions to the cumulated N<sub>2</sub>O flux over North America during 1979–2010 at continental and country levels

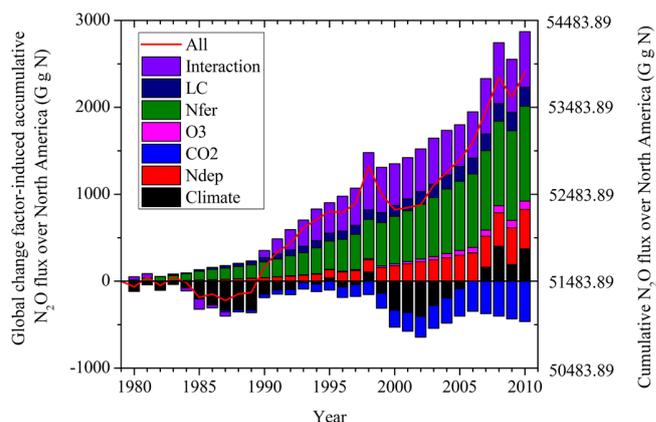
To quantify the relative contributions of multiple global change factors to the N<sub>2</sub>O flux over North America during 1979–2010, we summed up the individual factor-induced changes in N<sub>2</sub>O flux over the 32 yr to analyze the contributions of the six single factors and their interaction. To quantify the uncertainties associated with the cumulated N<sub>2</sub>O flux caused by the six individual factors and their interaction, we treated the thirty two annual fluxes as a sample to calculate the average flux and its standard error. Finally, the 32-yr cumulated flux and its standard error over the study period were reported. During the 32-yr study period, the cumulative N<sub>2</sub>O flux over North America was  $53.89 \pm 0.85$  Tg N<sub>2</sub>O-N, of which  $51.48 \pm 0.58$  Tg N<sub>2</sub>O-N was resulted from baseline flux and  $2.41 \pm 0.99$  Tg N<sub>2</sub>O-N was caused by global change factors (Fig. 5). Elevated atmospheric CO<sub>2</sub> decreased the N<sub>2</sub>O emission by  $0.46 \pm 0.06$  Tg N<sub>2</sub>O-N from North America's terrestrial ecosystems, while all the other single factors increased N<sub>2</sub>O emission. Climate variability, N deposition, O<sub>3</sub> pollution, N fertilizer application, land conversion, and multi-factor interaction enhanced continental N<sub>2</sub>O emission (Table 4).

To examine the factorial contributions to cumulated terrestrial N<sub>2</sub>O flux for the three countries, we further partitioned continental level N<sub>2</sub>O flux into country-level fluxes (Fig. 6). The 32-yr cumulative N<sub>2</sub>O emission was  $32.29 \pm 0.58$  Tg N<sub>2</sub>O-N for USA,  $9.38 \pm 0.22$  Tg N<sub>2</sub>O-N for Canada, and  $12.21 \pm 0.37$  Tg N<sub>2</sub>O-N for Mexico, respectively (Table 4). For the USA, climate variability and elevated atmospheric CO<sub>2</sub> decreased N<sub>2</sub>O emission, while N deposition, O<sub>3</sub> pollution, N fertilizer application, land conversion, and multi-factor interaction increased the country-level N<sub>2</sub>O emission (Table 4). After removing the baseline emission, the global change factors increased N<sub>2</sub>O emission from USA's terrestrial ecosystems from 1979 to 2010. For Canada, climate variability, N deposition, O<sub>3</sub> pollution, N fertilizer application, and land conversion increased N<sub>2</sub>O emission during 1979–2010; while elevated atmospheric CO<sub>2</sub> and multi-factor interaction decreased N<sub>2</sub>O emission (Table 4). After removing the baseline emission, the global change factors increased N<sub>2</sub>O emission from Canada's terrestrial ecosystems over the 32-yr period. For Mexico, climate variability, N deposition, O<sub>3</sub> pollution, N fertilizer application, and multi-factor interaction enhanced N<sub>2</sub>O emission during the time period of 1979–2010; while elevated atmospheric CO<sub>2</sub> and land conversion decreased N<sub>2</sub>O emission (Table 4). After removing the baseline flux, the global change factors stimulated N<sub>2</sub>O emission from Mexico's terrestrial ecosystems from 1979 to 2010.

For the country-level cumulative terrestrial N<sub>2</sub>O fluxes, the baseline emission made the biggest contribution: it accounted for 95.54 % of the continental N<sub>2</sub>O emission, and 97.18 %, 94.37 %, and 92.09 % of the N<sub>2</sub>O flux in the USA, Canada, and Mexico, respectively (Table 4).

### 3.6 Factorial contributions to the inter-annual variations in N<sub>2</sub>O flux over North America during 1979–2010 at continental and country levels

Inter-annual variation is one of major attributes of ecosystem processes; it may be caused by internal mechanisms or external environmental controls. Inter-annual variation in terrestrial N<sub>2</sub>O flux was shown over North America from 1979 to 2010 (Fig. 7). To examine the controlling factors for this inter-annual variation, we further attributed the changes in terrestrial N<sub>2</sub>O flux for each year to the six global change factors and their interaction (results shown in Fig. 7). Over the entire North America, rising atmospheric CO<sub>2</sub> continuously decreased, while N deposition and N fertilizer application continuously increased terrestrial N<sub>2</sub>O emission. O<sub>3</sub> pollution yielded very small positive effects on terrestrial N<sub>2</sub>O emission, while land conversion yielded small yet fluctuating effects on terrestrial N<sub>2</sub>O emission (Fig. 7). The climate variability primarily dominated the inter-annual fluctuation in terrestrial N<sub>2</sub>O flux from 1979 to 2010. Climate variability and multi-factor interaction co-dominated the increases in N<sub>2</sub>O emission in three specific time periods: 1979–1987,



**Fig. 5.** Factorial contributions to cumulated N<sub>2</sub>O flux over North America during 1979–2010 (The right y-axis shows the cumulative N<sub>2</sub>O flux with baseline; *All* means N<sub>2</sub>O flux derived from all combined simulation; *Interaction* means contribution from multi-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilizer application; *O<sub>3</sub>* means contribution from tropospheric O<sub>3</sub> pollution; *CO<sub>2</sub>* means contribution from elevated atmospheric CO<sub>2</sub>; *Ndep* means contribution from N deposition; *Climate* means contribution from climate variability).

1990–1998, and 2001–2008. For the specific years, such as 1988, 1989, and 1999 when the climatic contribution to the N<sub>2</sub>O flux was neutral, the interactive effect among multiple factors dominated the changes of terrestrial N<sub>2</sub>O flux compared to the baseline flux (Fig. 7).

After partitioning continental flux into country-level fluxes of N<sub>2</sub>O, we further analyzed and identified the major factors controlling the inter-annual fluctuation in terrestrial N<sub>2</sub>O over each country. It was found that the major factors leading to inter-annual fluctuation in terrestrial N<sub>2</sub>O flux varied among countries. Climate variability and multi-factor interaction co-dominated the inter-annual fluctuations in terrestrial N<sub>2</sub>O flux over the USA. During the periods of 1979–1986, 1990–1998, and 2009, the climatic effects dominated the terrestrial N<sub>2</sub>O emission over the USA. Meanwhile, interactive effects among the factors contributed to the flux of terrestrial N<sub>2</sub>O over the entire study period except the years of 2002–2003 (Fig. 8a).

Climate variability outweighed other factors in controlling the increases in terrestrial N<sub>2</sub>O emission over Canada during most of the study period (Fig. 8b). For instance, the climate-induced increases in terrestrial N<sub>2</sub>O flux were much higher than contributions from other factors during 2003–2010. However, in the years of 1982, 1989, 1992, 1995, 1998, and 2002, the climate-induced increases in terrestrial N<sub>2</sub>O were offset by the other factors' effects. Similar to the USA, the rising atmospheric CO<sub>2</sub> continuously decreased N<sub>2</sub>O emission. O<sub>3</sub> pollution, N input, and land conversion exerted minor effects on terrestrial N<sub>2</sub>O flux. The multi-factor interaction yielded very complex effects: it varied significantly through the study period, positive or negative,

**Table 4.** Factorial contributions to the cumulated N<sub>2</sub>O from 1979 to 2010 (*Climate* represents the impacts of climate variability only; *Ndep* represents the impacts of N deposition; *CO<sub>2</sub>* represents the impacts of CO<sub>2</sub> variation; *O<sub>3</sub>* represents the impacts of Tropospheric O<sub>3</sub> pollution; *Nfer* represents the impacts of N fertilizer application; *Land conversion* represents the impacts of land cover change only; *Interaction* represents the interactive effects of the six environmental factors).

	Baseline	Climate	Ndep	CO <sub>2</sub>	O <sub>3</sub>	Nfer	Land conversion	Interaction	Total flux
USA									
Cumulative N <sub>2</sub> O flux (T g N)	31.38 ± 0.45	-0.42 ± 0.48	0.20 ± 0.02	-0.17 ± 0.03	0.08 ± 0.01	0.84 ± 0.09	0.26 ± 0.02	0.12 ± 0.36	32.29 ± 0.58
Percentage (%)	97.18	-1.41	0.61	-0.53	0.24	2.60	0.82	0.38	100
Canada									
Cumulative N <sub>2</sub> O flux (T g N)	8.85 ± 0.12	0.75 ± 0.33	0.03 ± 0.005	-0.21 ± 0.02	0.004 ± 0.001	0.09 ± 0.01	0.007 ± 0.002	-0.14 ± 0.08	9.38 ± 0.22
Percentage (%)	94.37	7.98	0.34	-2.27	0.04	0.93	0.07	-1.47	100
Mexico									
Cumulative N <sub>2</sub> O flux (T g N)	11.25 ± 0.35	0.05 ± 0.41	0.22 ± 0.04	-0.08 ± 0.01	0.01 ± 0.003	0.16 ± 0.01	-0.05 ± 0.01	0.65 ± 0.13	12.21 ± 0.37
Percentage (%)	92.09	0.38	1.83	-0.67	0.11	1.34	-0.43	5.34	100
NA									
Cumulative N <sub>2</sub> O flux (T g N)	51.48 ± 0.58	0.37 ± 0.67	0.45 ± 0.06	-0.46 ± 0.06	0.10 ± 0.02	1.09 ± 0.11	0.22 ± 0.02	0.64 ± 0.39	53.89 ± 0.85
Percentage (%)	95.54	0.69	0.84	-0.86	0.18	2.02	0.41	1.19	100

Country- or individual factor-based estimates may not sum to totals because of the effects of rounding in reporting those estimates; positive values represent stimulating effects of global change factors on N<sub>2</sub>O flux; negative values represent inhibiting effects of global change factors on N<sub>2</sub>O flux.

small or large. For Mexico, although climate variability yielded significant effects on inter-annual variations in terrestrial N<sub>2</sub>O flux, the contributions from multi-factor interaction were predominated in several time periods. For example, in 2005, the multi-factor interaction outweighed other factors and played an important role in controlling terrestrial N<sub>2</sub>O flux; for the time period of 1983–1993, the contributions from multi-factor interaction also prevailed. The contributions from all other factors were in small magnitude (Fig. 8c).

## 4 Discussion

### 4.1 Comparisons with other studies

We compared our modeled results against previous studies to evaluate the factorial effects on N<sub>2</sub>O flux for major biomes (Table 5). Our estimated continental-scale average response to double CO<sub>2</sub> was -36.33% for forest ecosystems, which is consistent with Phillips et al.'s report that the N<sub>2</sub>O emission will be suppressed by elevated CO<sub>2</sub> (Phillips et al., 2001). DLEM simulated a 17.54% decrease of N<sub>2</sub>O emission from grassland, which is consistent with a previous estimate (Baggs et al., 2003). All field observations reported that the effects of elevated CO<sub>2</sub> on N<sub>2</sub>O emission depend on N availability; this could be explained by N limitation theory (Luo et al., 2004). Several studies have concluded that majorities of forest and grassland on the planet are N limited (Aber et al., 1998; Aber and Melillo, 2001; Vitousek

and Farrington, 1997; Vitousek et al., 1997), so the elevated CO<sub>2</sub> might suppress N<sub>2</sub>O emission from these ecosystems. Our study found that the elevated CO<sub>2</sub> suppressed N<sub>2</sub>O emission over North America's forest and grassland. This study also found that N deposition could stimulate N<sub>2</sub>O emission in all major biomes, which is consistent with field observations (Liu and Greaver, 2009; Ambus and Robertson, 2006; Kettunen et al., 2005). DLEM simulations indicate that N deposition could stimulate N<sub>2</sub>O emission at rates of 28.55 ± 0.24 mg N m<sup>-2</sup> a<sup>-1</sup>/(g N m<sup>-2</sup> a<sup>-1</sup>) for forest, 49.15 ± 0.19 mg N m<sup>-2</sup> a<sup>-1</sup>/(g N m<sup>-2</sup> a<sup>-1</sup>) for grassland, 47.77 ± 0.88 mg N m<sup>-2</sup> a<sup>-1</sup>/(g N m<sup>-2</sup> a<sup>-1</sup>) for wetland, and 13.94 ± 0.10 mg N m<sup>-2</sup> a<sup>-1</sup>/(g N m<sup>-2</sup> a<sup>-1</sup>) for cropland, respectively. These responses are larger than the summarized values by Liu and Greaver (2009). This might be related to the fact that our simulation study covered more heterogeneous space than Liu and Greaver (2009) did. There is only one reported field study for the effect of O<sub>3</sub> pollution on N<sub>2</sub>O emission from grassland (Kanerva et al., 2007). Both DLEM-estimation and field observation showed very small, or nearly neutral effect of O<sub>3</sub> pollution. O<sub>3</sub> has been found to be an inhibitor of plant growth (Ashmore, 2005; Kanerva et al., 2007; Wang et al., 2007); worse plant growth means lower N uptake, which will, in turn, increase N availability in soil and thus stimulate N<sub>2</sub>O emission.

A newly developed country-level inventory data of N<sub>2</sub>O fluxes was reported by United Nations Framework Convention on Climate Change (UNFCCC) (<http://unfccc.int>) in late 2011. A comparison shows that results from UNFCCC

**Table 5.** Comparison of factorial effects on N<sub>2</sub>O fluxes against other studies (positive values mean increase; negative values mean decrease in N<sub>2</sub>O emission).

	Biome	Experiment design	This study	Other studies	Literature
Elevated CO <sub>2</sub>	Forest	Double CO <sub>2</sub>	−36.33 %	Negative, neutral or positive effects, depending on seasons and nitrogen availability	Ambus and Robertson (1999); Phillips et al. (2001)
	Grassland	600 ppm vs. 360 ppm CO <sub>2</sub>	−17.54 %	−5.65 % ~ −26.01 % for low nitrogen input; 1.43 % ~62.27 % for high nitrogen input	Baggs et al. (2003); Ineson et al. (1998); Kanerva et al. (2007); Ambus and Robertson (2006)
Nitrogen deposition	Forest		28.55 ± 0.24 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	6 ± 1 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	Liu and Greaver (2009)
	Grassland		49.15 ± 0.19 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	6 ± 1 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	Liu and Greaver (2009)
	Wetland		47.77 ± 0.88 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	36 ± 13 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	Liu and Greaver (2009)
	Cropland		13.94 ± 0.10 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> ) for nitrogen deposition; 18.23 ± 0.05 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> ) for nitrogen fertilizer application	9 ± 1 (mg N m <sup>2</sup> a <sup>−1</sup> /g N m <sup>2</sup> a <sup>−1</sup> )	Liu and Greaver (2009)
Tropospheric O <sub>3</sub> pollution	Grassland	40–50 ppb in Open-top chambers	1.5 %	Decrease yet not significantly	Kanerva et al. (2007)

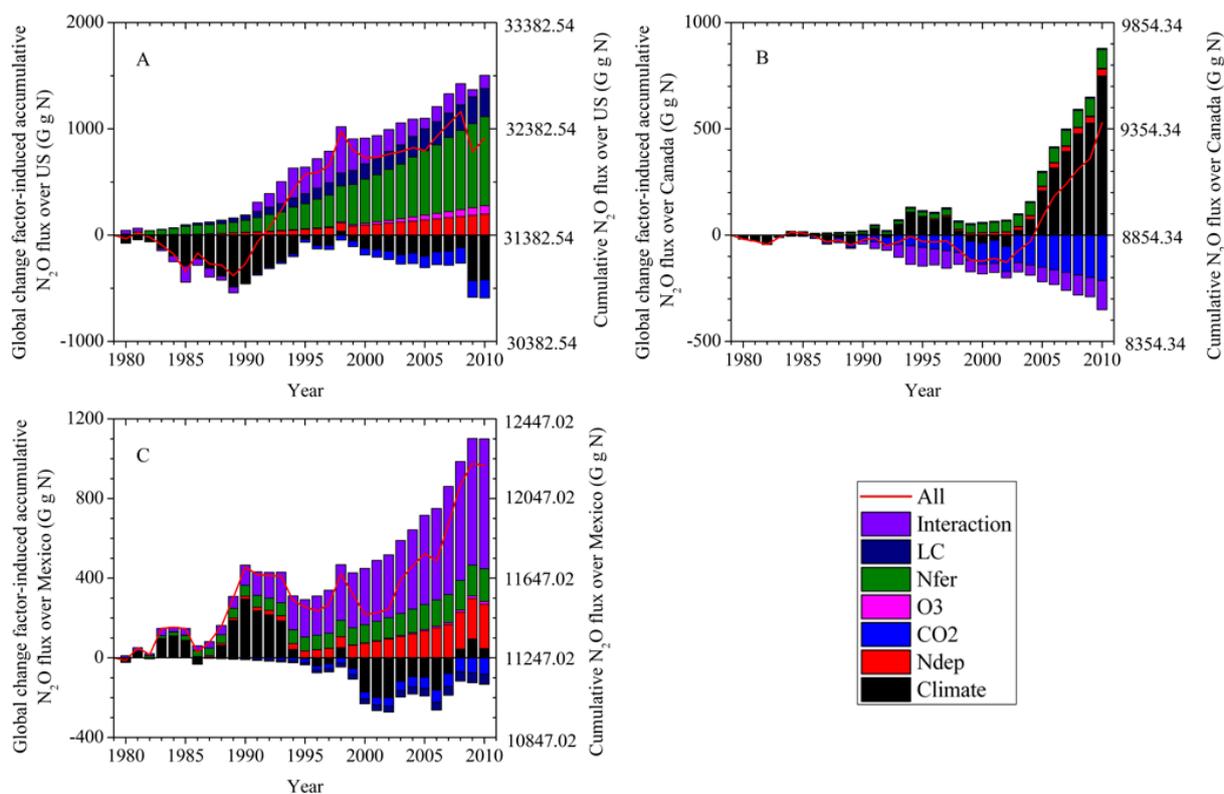
and this study are comparable yet different in magnitude due to different methods or datasets; for example, the UNFCCC estimates that N<sub>2</sub>O emission from agricultural soils in US is for 0.79 ~ 0.88 Tg N a<sup>−1</sup> from 1990 to 2009, while it is 0.35 ~ 0.44 Tg N a<sup>−1</sup> estimated by DLEM; this might be due to the fact that UNFCCC considers all agricultural land while DLEM only considers cropland.

#### 4.2 Factorial controls on N<sub>2</sub>O flux at temporal and spatial scales

Spatial variations in terrestrial N<sub>2</sub>O flux over North America simulated in this study were consistent with other studies (Xu et al., 2008; Potter et al., 1996). The major source for atmospheric N<sub>2</sub>O locates in southeastern continental North America, including the south part of central Canada, southeastern USA, and all of Mexico, which is associated with large cropland distributions and relatively high temperatures

(Fig. 3). The weak source in northern Canada is probably due to its low temperature and rainfall, as N<sub>2</sub>O emission is significantly controlled by temperature and soil moisture (Chapuis-Lardy et al., 2007; Conrad, 1996; Goldberg and Gebauer, 2009).

N<sub>2</sub>O flux was primarily controlled by environmental factors, substrate availability, and energy source (Brumme et al., 1999; Williams et al., 1992; Conrad, 1996). Global change factors alter terrestrial N<sub>2</sub>O flux through their effects on these processes (Dong et al., 2003; Kettunen et al., 2005; Mcswiney and Robertson, 2005; Kanerva et al., 2007; Zhang et al., 2007b). As consistent with previous studies (Mosier et al., 1991; Li et al., 1996; Mcswiney and Robertson, 2005; Zhang et al., 2007b), N input, including N deposition and N fertilizer application, enhanced the N<sub>2</sub>O emission from terrestrial ecosystems. O<sub>3</sub> has been confirmed to be a pollutant which may decrease productivity (Ashmore, 2005; Wang



**Fig. 6.** Factorial contributions to cumulated N<sub>2</sub>O flux at country-level during 1979–2010 (**A**: United States of America; **B**: Canada; **C**: Mexico) (The right y-axis shows the cumulative N<sub>2</sub>O flux with baseline; *All* means N<sub>2</sub>O flux derived from all combined simulation; *Interaction* means contribution from multi-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilizer application; *O<sub>3</sub>* means contribution from tropospheric O<sub>3</sub> pollution; *CO<sub>2</sub>* means contribution from elevated atmospheric CO<sub>2</sub>; *Ndep* means contribution from N deposition; *Climate* means contribution from climate variability).

et al., 2007), and thus suppress carbon source for microbial processes responsible for N<sub>2</sub>O production, and finally cause decreases in N<sub>2</sub>O emission. However, it is also reasonable to infer that the O<sub>3</sub> pollution inhibits productivity (Wang et al., 2007); and less N uptake might increase N availability in soil and in turn increase N<sub>2</sub>O emission. This study showed that the O<sub>3</sub> pollution enhanced N<sub>2</sub>O emission from terrestrial ecosystems in southeastern USA due to slightly higher N availability.

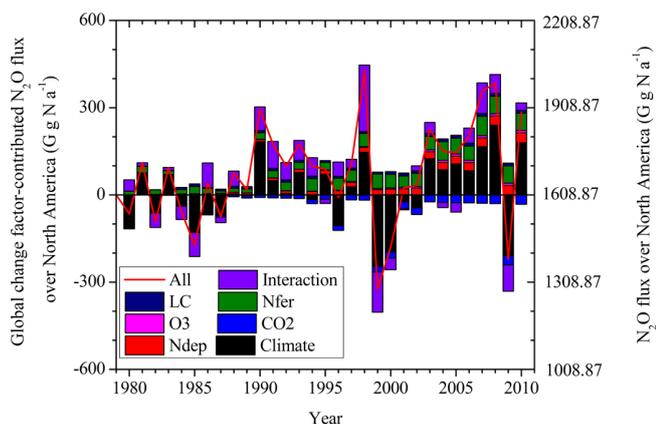
The elevated atmospheric CO<sub>2</sub> decreased N<sub>2</sub>O emission at continental scale; while the effects varied across North America. The stimulation effects of elevated atmospheric CO<sub>2</sub> on N<sub>2</sub>O emission were also observed over southeastern USA and portions of Mexico (Fig. 3c). In field experiments, both positive and negative effects of elevated atmospheric CO<sub>2</sub> on N<sub>2</sub>O emission were observed (Kammann et al., 2008; Phillips et al., 2001). The positive or negative effects might be determined by soil N availability; a field experiment concluded that a small amount of N fertilizer will relieve N limitation under elevated CO<sub>2</sub> concentration (Kettunen et al., 2007). If no progressive N limitation occurs under elevated CO<sub>2</sub>, enhanced N<sub>2</sub>O emission will be observed. As the theory of progressive N limitation predicts (Luo et al.,

2004), rising atmospheric CO<sub>2</sub> could lead to low N availability in soil and thus lead to low N<sub>2</sub>O emission (Kettunen et al., 2005; Barnard et al., 2004). In this study, the elevated atmospheric CO<sub>2</sub> substantially decreased the N<sub>2</sub>O emission from terrestrial ecosystem over North America, which is due to the N limitation for major biomes throughout the entire North America (Vitousek and Farrington, 1997; Aber and Melillo, 2001).

#### 4.3 Inter-annual variability in N<sub>2</sub>O flux

The increasing long-term trend and substantial inter-annual fluctuation in terrestrial N<sub>2</sub>O flux over North America during 1979–2010 resulted from complicated impacts from multiple factors (Chapuis-Lardy et al., 2007; Conrad, 1996; Ambus and Robertson, 1999). The highly varied climatic variability dominated the inter-annual flux of N<sub>2</sub>O over North America, and solely dominated the N<sub>2</sub>O flux over Canada. N deposition, O<sub>3</sub> pollution, and atmospheric CO<sub>2</sub> concentrations increased at relatively stable rates through the studied 32 yr, which resulted in the long-term trend of N<sub>2</sub>O flux (Fig. 8).

Using stepwise regression analysis, we found that climate variability was the predominating factor controlling



**Fig. 7.** Factorial contribution to the inter-annual variations in N<sub>2</sub>O flux over North America (The right y-axis shows the N<sub>2</sub>O flux with baseline; *All* means N<sub>2</sub>O flux derived from all combined simulation; *Interaction* means contribution from multi-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilizer application; *O<sub>3</sub>* means contribution from tropospheric O<sub>3</sub> pollution; *CO<sub>2</sub>* means contribution from elevated atmospheric CO<sub>2</sub>; *Ndep* means contribution from N deposition; *Climate* means contribution from climate variability).

inter-annual variations in terrestrial N<sub>2</sub>O flux at both continental and country levels. Following climate variability, multi-factor interaction played an essential role in contributing temporal variations in terrestrial N<sub>2</sub>O flux. This is consistent with our previous analysis showing that climate variability, followed by multi-factor interaction, co-dominated inter-annual variations in terrestrial N<sub>2</sub>O flux at both continental and country levels (Sect. 3.6). The importance of climate variability in controlling inter-annual N<sub>2</sub>O emission indicates the important role of climate variability in controlling variations in atmospheric N<sub>2</sub>O concentration (Fluckiger et al., 1999).

#### 4.4 Interactions among multiple factors

The interaction among global change factors has been long recognized as an important factor (Dermody, 2006). A large amount of field experiments still treat it as negligible, although a few experiments have introduced this in their experiment design (Xia et al., 2009; Reich et al., 2006). Through this study, the multi-factor interaction was recognized playing an important role in contributing to terrestrial N<sub>2</sub>O flux (Sect. 3.6). Since the various climate variables are associated in reality, the separation of each climate variable might be biased. Because the field experiments are usually labor intensive, multi-factor experiment might be hard to carry out; the interactive effects among more than three factors are still short of investigation (Heimann and Reichstein, 2008; Norby and Luo, 2004). This study shows that the modeling approach may serve as one complementary tool for field exper-

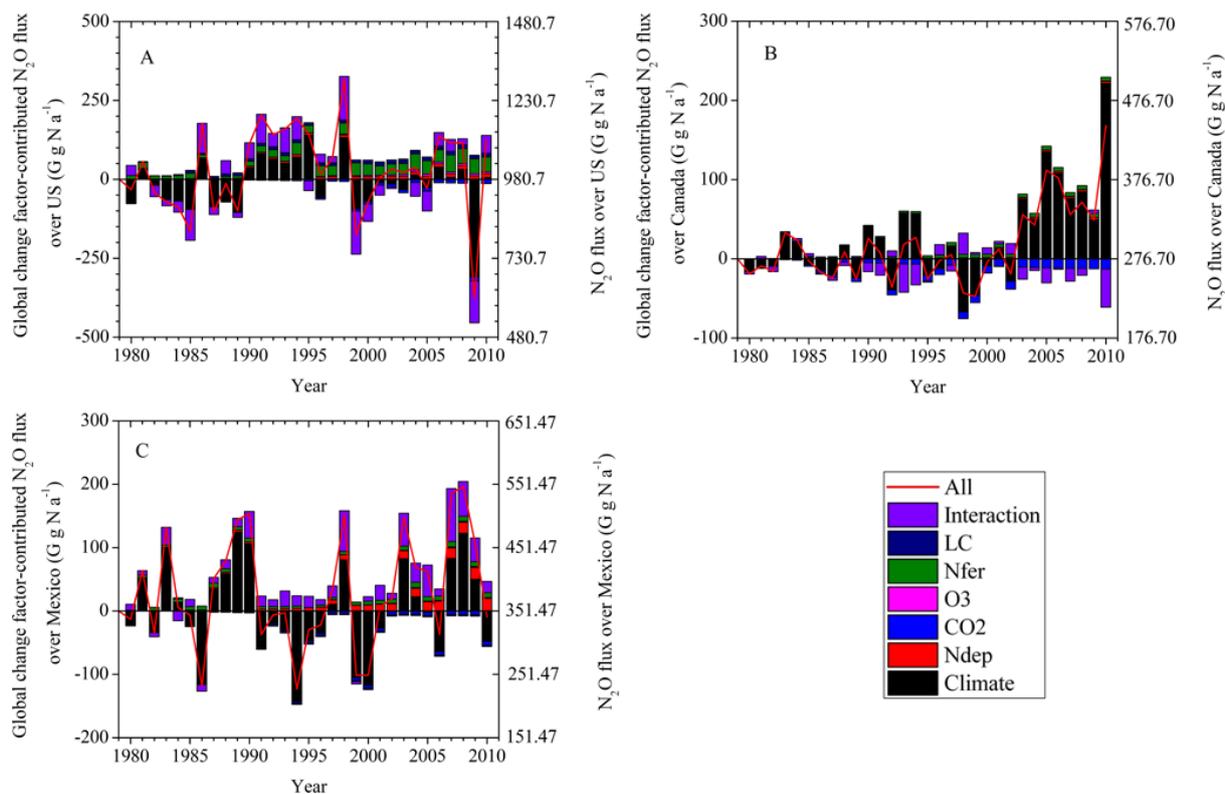
iments in addressing interactive effect in a multi-factor world (Norby and Luo, 2004).

#### 4.5 Uncertainties and future research needs

Considering the limitations in existing manipulative field experiments on N<sub>2</sub>O fluxes, and the complexities in the related nonlinear processes and multiple controlling factors, numerical experiments with models like DLEM that represent the fundamental mechanisms in N and N gas processes and the coupled biogeochemical cycles become an important approach to gain insights into the dynamics of N-containing gas emission in the context of global changes. This is the reason why process-based models are widely used in climate change studies, and why our study places so much emphasis on the relative contribution and interactions of multiple environmental controls. Through this study, rather than try to draw any conclusions on this issue, we hope to provide some insight on the patterns and dynamics of the N gas emission and to raise some questions about the effects of multiple environmental factors on the process.

This study was able to attribute the spatial and temporal variations in N<sub>2</sub>O flux over North America's terrestrial ecosystems during 1979–2010, but there are several issues that still need to be improved. First, this study only considered the land conversion between cropland and natural vegetation, and would generate more accurate results if other land use changes such as afforestation were included. Second, the pre-1979 legacy effect may cause some biases in this study, which solely analyzed N<sub>2</sub>O flux over the time period of 1979–2010. Third, the soil may act as a sink of atmospheric N<sub>2</sub>O (Chapuis-Lardy et al., 2007); however, owing to incomplete understanding of this phenomenon, we did not incorporate this mechanism in this study, so the model may have overestimated the terrestrial N<sub>2</sub>O flux at both continental and county levels. More field studies are needed to understand the mechanisms for N<sub>2</sub>O sink in soil, which will improve the regional estimation of N<sub>2</sub>O flux.

Fourth, although we have compared our estimated factorial impacts on N<sub>2</sub>O flux with other studies, the effects of global change factors on N<sub>2</sub>O have not been comprehensively calibrated and validated because of the scarcity of field observations on factorial N<sub>2</sub>O fluxes (Dermody, 2006). Fifth, the thawing-freezing cycle has long been identified as a major factor on N<sub>2</sub>O emission, especially in temperate and boreal regions (de Bruijn et al., 2009; Repo et al., 2009; Christensen and Tiedje, 1990), yet the real mechanisms are still not well investigated (de Bruijn et al., 2009). So this study might have underestimated the N<sub>2</sub>O emission due to lack of freezing-thawing effect. Sixth, although the regional data used in this study are fairly updated, improvements of the dataset might provide more accurate estimations. For instance, nitrogen deposition data was interpolated based on three annual maps of spatial distribution of nitrogen deposition (Dentener, 2006); improved atmospheric transport



**Fig. 8.** Factorial contribution to the inter-annual variations in N<sub>2</sub>O flux at country level (**A:** United States of America; **B:** Canada; **C:** Mexico) (The right y-axis shows the N<sub>2</sub>O flux with baseline; *All* means N<sub>2</sub>O flux derived from all combined simulation; *Interaction* means contribution from multi-factor interaction; *LC* means contribution from land conversion; *Nfer* means contribution from N fertilizer application; *O<sub>3</sub>* means contribution from tropospheric O<sub>3</sub> pollution; *CO<sub>2</sub>* means contribution from elevated atmospheric CO<sub>2</sub>; *Ndep* means contribution from N deposition; *Climate* means contribution from climate variability).

model associated with more field observations might provide more accurate nitrogen deposition data. Seventh, further work on N<sub>2</sub>O flux in cropland is needed to reduce the uncertainty because of the dominance of fertilized cropland as N<sub>2</sub>O source and the shortage of extensive validation of DLEM-simulated N<sub>2</sub>O flux. Last but not least, it will be an improvement when additional environmental factors such as wild and prescribed fires, harvests, insect outbreaks, etc. are considered. Meanwhile, we acknowledged that the precision is different from accuracy; although many simulations were performed and uncertainty analysis was conducted, more efforts are needed to increase the accuracy of estimated N<sub>2</sub>O fluxes.

## 5 Conclusions

This study examined the factorial contributions to the terrestrial N<sub>2</sub>O flux over North America at both continental and country levels by using a highly-integrated process-based ecosystem model driven by multiple global change factors, including changing climate, N deposition, rising atmospheric CO<sub>2</sub>, O<sub>3</sub> pollution, N fertilizer application, and land conver-

sion. The improvements in input data and model mechanisms are needed for more accurate estimates. Nevertheless, this study is helpful in advancing our understanding of the dynamics of atmospheric N<sub>2</sub>O concentration as well as beneficial for the policy-makers to curb the increase in atmospheric N<sub>2</sub>O concentration. The complicated effects of multi-factor interaction on N<sub>2</sub>O flux suggest that the current field experiments, which usually ignore the interactive effects from multiple factors, may lead to biases in the estimation of N<sub>2</sub>O flux. This study also pointed out that the models driven by only a few global change factors may bring bias in estimating N<sub>2</sub>O flux.

This study is among the first attempts to attribute the spatiotemporal variations in regional terrestrial N<sub>2</sub>O flux to multiple global change factors over a long time period. A collaborative effort between field ecologists and modelers is necessary for further investigation of the underlying mechanisms responsible for spatial and temporal variations in N<sub>2</sub>O flux.

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