

**Controls on
terrestrial N₂O flux in
North America**

X. F. Xu et al.

Multiple-factor controls on terrestrial N₂O flux over North America from 1979 through 2010

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

Nitrous oxide (N_2O) is a potent greenhouse gas which also contributes to the depletion of stratospheric ozone (O_3). However, the magnitude and underlying mechanisms for the spatiotemporal variations in the terrestrial sources of N_2O are still far from certain. Using a process-based ecosystem model (DLEM – the Dynamic Land Ecosystem Model) driven by multiple global change factors, including climate variability, nitrogen (N) deposition, rising atmospheric CO_2 , tropospheric O_3 pollution, N fertilizer application, and land conversion, the spatial and temporal variations in terrestrial N_2O flux over North America were examined and attributed to various driving factors. From 1979 to 2010, the North America accumulatively emitted $55.1 \pm 0.8 \text{ Tg } \text{N}_2\text{O-N}$ ($1 \text{ Tg} = 10^{12} \text{ g}$), of which global change factors contributed $2.8 \pm 1.0 \text{ Tg } \text{N}_2\text{O-N}$, and baseline emission contributed $52.3 \pm 0.6 \text{ Tg } \text{N}_2\text{O-N}$. Climate variability, N deposition, O_3 pollution, N fertilizer application, and land conversion increased N_2O emission by $0.3 \pm 0.7 \text{ Tg } \text{N}_2\text{O-N}$, $0.5 \pm 0.1 \text{ Tg } \text{N}_2\text{O-N}$, $0.11 \pm 0.02 \text{ Tg } \text{N}_2\text{O-N}$, $1.2 \pm 0.1 \text{ Tg } \text{N}_2\text{O-N}$, and $0.2 \pm 0.02 \text{ Tg } \text{N}_2\text{O-N}$, respectively. The elevated atmospheric CO_2 led to a decrease in terrestrial N_2O emission by $0.5 \pm 0.07 \text{ Tg } \text{N}_2\text{O-N}$. The interactive effect among multiple factors enhanced N_2O emission by $0.9 \pm 0.3 \text{ Tg } \text{N}_2\text{O-N}$ over the 32 years. At country level, climate variability and elevated atmospheric CO_2 decreased, while all other single factors and multiple-factor interaction enhanced N_2O emission in the United States of America (USA) over the study period. During the same time period, elevated atmospheric CO_2 and multiple-factor interaction decreased, while other factors enhanced N_2O emission from Canada. Elevated atmospheric CO_2 and land conversion decreased while other factors enhanced N_2O emission from Mexico. The interactive effects among climate variables play a predominant role in controlling climate-induced changes in N_2O emission at both continental and country levels. Central and southeastern parts of the North America – including central Canada, central USA, southeastern USA, and all of Mexico – experienced increases in N_2O emission from 1979 to 2010. The effects of climate variability and multiple-factor interaction dominating the inter-annual variations in

Controls on terrestrial N_2O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



terrestrial N₂O emission at both continental and country levels indicate that projected changes in the global climate system during this century may substantially alter the regime of N₂O emission from terrestrial ecosystems. They also imply that the interactive effect among global change factors may significantly affect N₂O flux, needing more investigations through field experiments.

1 Introduction

Nitrous oxide (N₂O) plays an important role in both contributing to the greenhouse effect (Denman et al., 2007; Rodhe, 1990) and depleting stratospheric ozone (O₃) (Denman et al., 2007; Cicerone, 1987). The atmospheric N₂O 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 N₂O flux and the underlying mechanisms will be crucial for advancing our understanding of the dynamics of atmospheric N₂O concentration, and further provide helpful information for policy-makers to curb the continuous increase in atmospheric N₂O concentration (Tian et al., 2010b; Denman et al., 2007).

N₂O 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 N₂O 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 N₂O production by increasing substrate availability (Kettunen et al., 2005; Mcswiney and Robertson, 2005); elevated atmospheric CO₂ 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 N₂O emission (Phillips et al., 2001); alternatively, elevated

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



atmospheric CO₂ might increase photosynthetic products and stimulate microbial process, and thus increase N₂O emission (Kettunen et al., 2005; Ineson et al., 1998). If these two effects are counterbalanced, it may appear as neutral response of N₂O flux to elevated atmospheric CO₂ (Kanerva et al., 2007; Ambus and Robertson, 1999). Tropospheric O₃ pollution may alter microbial community (Kanerva et al., 2008) and cause an increase or decrease in N₂O emission, depending on time and location (Kanerva et al., 2007). The effects of climate variability and land conversion on the N₂O emission are more complicated and are really dependent on 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 N₂O 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 N₂O 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 neglected, at least partially (Potter et al., 1996; Matson et al., 1989). Meanwhile, the empirical methods in estimating regional N₂O flux could not be used to attribute the spatiotemporal variations in terrestrial N₂O flux to underlying mechanisms. So, a large-scale estimation of terrestrial N₂O 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 conducting regional estimation of N₂O flux (Del Grosso et al., 2006; Li et al., 2001; Potter et al., 1996). Although the process-based models have the potential to explore the relative contributions from each driving force to the spatiotemporal variations in terrestrial N₂O, as they have been conducted for CO₂ flux (McGuire et al., 2001; Tian et al., 2003; Mu et al.,

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2008) and CH₄ flux (Xu et al., 2010), none have been utilized to attribute the spatial and temporal variations in terrestrial N₂O flux to its driving factors.

North America, one of the extensively investigated continents, still lacks accurate estimates due to limitations in method and observations (Kort et al., 2008). A number of studies estimated the N₂O 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 N₂O emission from cropland in the United State of America (USA) by using a process-based model DNDC (Li et al., 1996). Del Grosso et al. (2006) estimated the N₂O flux from cropland in the USA by using the DAYCENT model (Del Grosso et al., 2006). One of our previous studies simulated the regional flux of N₂O 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 CO₂, N deposition, O₃ pollution, N fertilizer application, and land use change (Tian et al., 2010b). In this study, we will extend our previous work and attribute the variations in terrestrial N₂O at spatial and temporal scales to various driving factors.

Specifically, the objectives of this study are: (1) to examine the factorial contributions to the spatial variation of terrestrial N₂O flux over North America during 1979–2010; (2) to quantify the factorial contributions to the temporal variations in terrestrial N₂O flux over North America during 1979–2010; (3) to quantify the factorial contributions to the 32-year accumulative flux of N₂O over North America at both continental and country levels; and (4) to identify the major factors responsible for the spatial and temporal variations in terrestrial N₂O flux at both continental and country levels. The global change factors evaluated in this study include climate variability, rising atmospheric CO₂, N deposition, O₃ pollution, changes in land use and land cover type, and N fertilizer application in cropland. The interactive effects among these six factors were calculated by subtracting the changes in N₂O flux contributed from all factors together by the changes in N₂O flux caused by six individual factors (see Experiment design section for the detail information).

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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, shrub, 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., 2011), and the N₂O module has been described in detail in Tian et al. (2010b).

In the DLEM, the N₂O 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 NH₄⁺ concentration (Lin et al., 2000; Tian et al., 2010b). 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 NO₃⁻ concentration (Lin et al., 2000). The empirical equation reported by

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Davidson et al. (2000) is used to separate N₂O from other gases (mainly NO and N₂).

In summary, multiple global change factors directly and/or indirectly affect N₂O processes in the DLEM (Fig. 1), which could be expressed as the following equation:

$$N_2O = V_{\max} f(C_a, w_j, T_{\text{air}}, \text{APAR}) f(O_3) f(N) \quad (1)$$

5 where N₂O is the N₂O flux; V_{max} is the maximum rate of N₂O production via nitrification and denitrification; $f(C_a, w_j, T_{\text{air}}, \text{APAR})$ describes the effects of air temperature, atmospheric CO₂ concentration, soil moisture, and absorbed photosynthetically active radiation on N₂O flux through its photosynthesis; $f(O_3)$ describes the effects of O₃ pollution; $f(N)$ describes the effect of N input; C_a is atmospheric CO₂ concentration, w_j is soil moisture; T_{air} is daily temperature, APAR is absorbed photosynthetically active radiation. It should be noted that the above equation solely includes the direct and indirect effects from multiple global change factors on N₂O flux; the other environmental factors that might influence N₂O flux were not incorporated in the equation, for example, soil pH and soil porosity, etc.

15 2.2 Study area and input data

North America was selected in this study. It includes the USA, Canada, and Mexico, covering a total area of approximately 24.71 million km², which is about 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 resolution of 32 km by 32 km, which is consistent with the North American Regional Reanalysis (NARR) dataset.

20 We developed gridded (32 km × 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

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



NARR dataset (Mesinger et al., 2006). The land use and land cover change data, N fertilizer data, O₃ pollution data, and N deposition were assumed unchanged after 2005. The annual atmospheric concentration of CO₂ was updated based on the dataset from National Oceanic and Atmospheric Administration (NOAA) (www.esrl.noaa.gov).

Historical data from 1901 to 2010 are prescribed as transient input data sets in this study. The transient input 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 O₃ pollution data from 1901 to 2010; (4) atmospheric CO₂ 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

Similar to one of our previous studies (Xu et al., 2010), we conducted nineteen simulations in this study to determine the relative effects of N deposition, O₃ pollution, climate variability, elevated atmospheric CO₂, land-use change, and N fertilizer application on terrestrial N₂O flux over North America. One overall simulation was set up to simulate the terrestrial N₂O flux over North America by considering the temporal and spatial dynamics of all six global change factors. Six simulations were set up to simulate the effects of each individual factor on N₂O 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 in 1900: the atmospheric CO₂ concentration, N deposition, O₃ 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 CO₂ fertilization alone, we ran DLEM using the historical atmospheric CO₂ concentrations, but kept all other

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



five global change factors constant: a 32-year average daily climate data was used to represent the mean climate data, the potential vegetation map with crop and urban land in 1900 was used to represent the constant land cover type, and the N deposition, O₃ pollution, and N fertilizer application data were kept constant at the year of 1900.

5 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 design was used to capture the internal dynamics of the system, which served as baseline.

Five simulations were set up to separate the contributions from each single climate variable: precipitation, temperature (maximum, average, minimum), solar radiation, and relative humidity. Four simulations were set up to simulate the contribution from 10 each of four climate variables, and one was set up as baseline to exclude the contribution from system dynamics; i.e. the post-1979 simulations were fed by 1979 climate data (Table 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 15 vegetation, long-term mean climate during 1979–2010, the concentration levels of N deposition, O₃, and atmospheric CO₂ in the year 1900 to drive the model simulations to an equilibrium state (i.e. the inter-annual variations are $< 0.1 \text{ g C m}^{-2}$ for carbon storage, $< 0.1 \text{ g N m}^{-2}$ for N storage). After the system reached an equilibrium state, the 20 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 CO₂ concentration, and N deposition inputs from 1901 to 2010 to simulate the terrestrial N₂O flux. Only the outputs between 1979 and 2010 were analyzed to show the spatial and temporal patterns of terrestrial N₂O flux in North America's 25 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 N₂O flux during 1979–2010 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.

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



2.4 Model parameterization

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

2.5 Statistic method

The regression analysis was used in this study to find the long-term changing trend of input data and N₂O fluxes generated by various simulations. All the statistic 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 $0.04 \pm 0.01 \text{ }^\circ\text{C a}^{-1}$, $0.03 \pm 0.01 \text{ }^\circ\text{C a}^{-1}$, $0.03 \pm 0.01 \text{ }^\circ\text{C a}^{-1}$, 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 O₃ 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 CO₂ concentration, respectively (Table 2). The areas of different land cover types changed slightly through the study period: the cropland area increased from 2.51 million km² to 2.59 million km²; the areas of forest, shrub, grassland, and wetland changed in very small magnitudes (Table 3).

Spatial variations of input data were shown in a previous paper (Xu et al., 2010). Normally, the severely O₃-polluted areas over North America locate in western parts

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of North America and the southeastern USA, which could be as high as 5000 ppb h⁻¹ (monthly accumulated hourly O₃ dose over a threshold of 40 ppb in ppb-h); while the other areas, especially the northern end of continental North America, feature low O₃ pollution. The regions with high N fertilizer application (larger than 10 g N m⁻² a⁻¹) concentrate in the USA, including western, central, and eastern coastal areas of the USA. Canada and Mexico had smaller amounts of cropland and received lower application rates of N fertilizer. The high N deposition primarily occurred in eastern parts of continental North America, including southeastern Canada, eastern USA, and portions of Mexico (larger than 1 g N m⁻² a⁻¹), while northern Canada featured quite low N deposition (lower than 0.01 g N m⁻² a⁻¹).

3.2 Spatial distribution of N₂O flux over North America during 1979–2010

The terrestrial N₂O 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 N₂O emission, up to nearly 1 g N m⁻² a⁻¹, while western USA had relatively low N₂O emission rates. There was a north-to-south increasing gradient of N₂O emission across Canada: the N₂O emission rate was as low as 0.0001 g N m⁻² a⁻¹ in northern Canada, and as high as nearly 0.4 g N m⁻² a⁻¹ in southern Canada. The entire country of Mexico acted as a strong source for N₂O, with the national N₂O emission being 0.16 ± 0.03 g N m⁻² a⁻¹ over the 32-year period.

3.3 Factorial contribution to the spatial variations in terrestrial N₂O flux during 1979–2010

Nitrogen release as N₂O is one of major pathways for nitrogen loss from terrestrial ecosystem. When external stresses are posed upon, the terrestrial N₂O emission will be changed. The terrestrial N₂O flux with no external stress is defined as background

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



emission; the changes in terrestrial N₂O flux caused by external factors are external-factor-induced N₂O flux. Throughout the study period, the accumulative N₂O emission over North America was composed of two components: one is the background emission, defined as the N₂O flux during 1979–2010 simulated by the DLEM driven by the input data of 1979; the other is the flux contributed from changes in various individual and interactive effects of several global change factors. After removing the baseline flux of N₂O, the changes in N₂O fluxes over the study period were resulted from six global change factors and their interaction.

Over the 32-year study period, climate variations enhanced N₂O emission over the majority of central Canada, while decreased N₂O emissions over portions of western Alaska, majority of the USA, and a portion of Mexico (Fig. 3a). N deposition enhanced N₂O emission over the entire continental North America, with prominent increases over southeastern USA and Mexico (Fig. 3b). The elevated atmospheric CO₂ enhanced N₂O emissions in southeastern USA and a portion of Mexico, while decreased N₂O emissions in western USA, central Canada, and the majority of Mexico (Fig. 3c). The O₃ pollution increased N₂O emissions in portions of southeastern USA, while no significant impacts on N₂O flux were yielded in other areas (Fig. 3d). The N fertilizer application enhanced N₂O emission in central USA, which is agricultural land (Fig. 3e), while 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 N₂O emissions in central USA, while decreased N₂O emissions in southeastern USA and central Canada (Fig. 3h). Combining all the effects from various global change factors, the N₂O 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 N₂O flux over North America during 1979–2010

The continental-level terrestrial flux of N₂O over the North America showed a significant inter-annual fluctuation during 1979–2010 (Fig. 4). The mean annual N₂O

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



flux over North America's terrestrial ecosystems was $1.72 \pm 0.15 \text{ Tg N}_2\text{O-N a}^{-1}$, with an overall increasing rate of $6.16 \pm 2.66 \text{ Gg N}_2\text{O-N a}^{-1}$ ($1 \text{ Gg} = 10^9 \text{ g}$) over 32 years ($P = 0.028$). The long-term increasing trend and inter-annual fluctuation in terrestrial N_2O flux were resulted from multiple global change factors (Fig. 4). Climate variability exerted a significant inter-annual variation in N_2O flux, with a slightly increasing rate of $2.86 \pm 2.20 \text{ Gg N}_2\text{O-N a}^{-1}$ ($P = 0.204$). N deposition contributed to an increasing rate of $1.49 \pm 0.02 \text{ Gg N}_2\text{O-N a}^{-1}$ ($P < 0.001$), and N fertilizer application contributed to an increasing rate at $2.45 \pm 0.10 \text{ Gg N}_2\text{O-N a}^{-1}$ for continental N_2O emission ($P < 0.001$). O_3 pollution contributed to an increasing rate of $0.33 \pm 0.03 \text{ Gg N}_2\text{O-N a}^{-1}$ ($P < 0.001$). The increased atmospheric CO_2 concentration continuously decreased N_2O emission, which generated a decreasing rate of $1.79 \pm 0.07 \text{ Gg N}_2\text{O-N a}^{-1}$ ($P < 0.001$) over the study period, while land conversion did not yield significant changing trends of N_2O emission over the study period.

3.5 Factorial contributions to the accumulated N_2O flux over North America during 1979–2010 at continental and country levels

To quantify the relative contributions from multiple global change factors to the N_2O flux over North America during 1979–2010, we summed up the individual factor-induced changes in N_2O flux over the 32 years to analyze the contributions of the six single factors and their interaction. To express the uncertainties associated with the accumulated N_2O 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-year accumulated flux and its standard error over the study period were reported. Through the 32-year study period, the accumulative N_2O flux over North America was $55.08 \pm 0.84 \text{ Tg N}_2\text{O-N}$, of which $52.32 \pm 0.57 \text{ Tg N}_2\text{O-N}$ was contributed from baseline flux and $2.76 \pm 0.95 \text{ Tg N}_2\text{O-N}$ was caused by global change factors. Elevated atmospheric CO_2 decreased the N_2O emission by $0.52 \pm 0.07 \text{ Tg N}_2\text{O-N}$ from North America's terrestrial ecosystems, while all the other single factors increased N_2O

BGD

8, 10935–10977, 2011

Controls on terrestrial N_2O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



emission. Climate variations, N deposition, O₃ pollution, N fertilizer application, land conversion, and multiple-factor interaction enhanced continental N₂O emission (Table 4).

To examine the factorial contributions to accumulated terrestrial N₂O flux for the three countries, we further partitioned continental level N₂O flux into country-level fluxes. The 32-year accumulative N₂O emission was 33.49 ± 0.57 Tg N₂O-N for USA, 9.38 ± 0.22 Tg N₂O-N for Canada, and 12.21 ± 0.37 Tg N₂O-N for Mexico, respectively (Table 4). For the USA, climate variability and elevated atmospheric CO₂ decreased N₂O emission, while N deposition, O₃ pollution, N fertilizer application, land conversion, and multiple-factor interaction increased the country-level N₂O emission (Table 4). After removing the baseline emission, the global change factors increased N₂O emission from USA's terrestrial ecosystems from 1979 to 2010. For Canada, climate variations, N deposition, O₃ pollution, N fertilizer application, and land conversion increased N₂O emission during 1979–2010; while elevated atmospheric CO₂ and multiple-factor interaction decreased N₂O emission (Table 4). After removing the baseline emission, the global change factors increased N₂O emission from Canada's terrestrial ecosystems over the 32-year period. For Mexico, climate variability, N deposition, O₃ pollution, N fertilizer application, and multiple-factor interaction enhanced N₂O emission during the time period of 1979–2010; while elevated atmospheric CO₂ and land conversion decreased N₂O emission (Table 4). After removing the baseline flux, the global change factors stimulated N₂O emission from Mexico's terrestrial ecosystems from 1979 to 2010.

For the country-level accumulative terrestrial N₂O fluxes, the baseline emission made the biggest contribution: it accounted for 94.99 % of the continental N₂O emission, and 96.22 %, 94.37 %, and 92.09 % of the N₂O flux in the USA, Canada, and Mexico, respectively (Table 4).

Since four climate variables including precipitation, relative humidity, solar radiation, and temperature have been considered in this study, we further conducted simulations to estimate the contributions of each climate variable to the changes in climate-induced

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



N₂O fluxes at country and continental levels (Table 5). All single variables made negative effects on N₂O emission in the USA and over continental North America, and all variables except precipitation made negative effects on N₂O emission in Canada, while all climate variables except temperature made negative effects on N₂O emission in Mexico. Multiple variables interaction was predominant in controlling N₂O emission at both country and continental scales, indicating the inter-correlation among climate variables.

3.6 Factorial contributions to the inter-annual variations in N₂O 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 control. Inter-annual variation in terrestrial N₂O 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 N₂O 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 CO₂ continuously decreased, while N deposition and N fertilizer application continuously increased terrestrial N₂O emission. O₃ pollution yielded very small positive effects on terrestrial N₂O emission, while land conversion yielded small yet fluctuating effects on terrestrial N₂O emission (Fig. 7). The climate variability primarily dominated the inter-annual fluctuation in terrestrial N₂O flux from 1979 to 2010. Climate variability and multiple-factor interaction co-dominated the increases in N₂O emission in three specific time periods: 1979–1987, 1990–1998, and 2001–2008. For the specific years, such as 1988, 1989, and 1999 when the climatic contribution to the N₂O flux was neutral, the interactive effect among multiple factors dominated the changes of terrestrial N₂O flux compared to the baseline flux (Fig. 7).

After partitioning continental flux into country-level fluxes of N₂O, we further analyzed and identified the major factors controlling the inter-annual fluctuation in terrestrial N₂O over each country. It was found that the major factors leading to inter-annual fluctuation

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



in terrestrial N₂O flux varied over different countries. Climate variability and multiple-factor interaction co-dominated the inter-annual fluctuations in terrestrial N₂O flux over the USA. During the periods of 1979–1986, 1990–1998, and 2009, the climatic effects dominated the terrestrial N₂O emission over the USA. Meanwhile, interactive effects among the factors contributed to the flux of terrestrial N₂O over the entire study period except the years of 2002–2003 (Fig. 8a).

Climate variability outweighed other factors in controlling the increases in terrestrial N₂O emission over Canada during most of the study period (Fig. 8b). For instance, the climate-induced increases in terrestrial N₂O 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 N₂O were offset by the other factors' effects. Similar to the USA, the rising atmospheric CO₂ continuously decreased N₂O emission. O₃ pollution, N input, and land conversion exerted minor effects on terrestrial N₂O flux. The multiple-factor interaction yielded very complex effects: it varied significantly through the study period, positive or negative, small or large. For Mexico, although climate variability yielded significant effects on inter-annual variations in terrestrial N₂O flux, the contributions from multiple-factor interaction were predominated in several time periods. For example, in 2005, the multiple-factor interaction outweighed other factors and played an important role in controlling terrestrial N₂O flux; for the time period of 1983–1993, the contributions from multiple-factor interaction also prevailed. The contributions from all other factors were in small magnitude (Fig. 8c).

4 Discussion

4.1 Comparison with other studies

We compared our modeled results against previous studies to evaluate the factorial effects on N₂O flux for major biomes (Table 6). Our estimated continental-level average

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



response to double CO₂ was -36.33% for forest ecosystems, which is consistent with Phillips et al.'s report that the N₂O emission will be suppressed by elevated CO₂ (Phillips et al., 2001). DLEM simulated a 17.54% decrease of N₂O emission from grassland, which is consistent with a previous estimate (Baggs et al., 2003). All field observations reported that the effects of elevated CO₂ on N₂O 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 CO₂ might suppress N₂O emission from these ecosystems. Our study found that the elevated CO₂ suppressed N₂O emission over North America's forest and grassland. This study also found that N deposition could stimulate N₂O 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 N₂O emission at rates of $28.55 \pm 0.24 \text{ mg N m}^{-2} \text{ a}^{-1} / (\text{g N m}^{-2} \text{ a}^{-1})$ for forest, $49.15 \pm 0.19 \text{ mg N m}^{-2} \text{ a}^{-1} / (\text{g N m}^{-2} \text{ a}^{-1})$ for grassland, $47.77 \pm 0.88 \text{ mg N m}^{-2} \text{ a}^{-1} / (\text{g N m}^{-2} \text{ a}^{-1})$ for wetland, and $13.94 \pm 0.10 \text{ mg N m}^{-2} \text{ a}^{-1} / (\text{g N m}^{-2} \text{ a}^{-1})$ for cropland, respectively. These responses are larger than the summarized values by Liu and Greaver (2009). This might be due to the lack of data collection in Liu and Greaver (2009) or others, which are worthy of more investigation. There is only one reported field study for the effect of O₃ pollution on N₂O emission from grassland (Kanerva et al., 2007). The DLEM-estimated result is different from field observation, yet both reported responses are very small, nearly neutral effects. O₃ 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 N₂O emission.

4.2 Factorial controls on N₂O flux at temporal and spatial scales

Spatial variations in terrestrial N₂O flux over North America simulated in this study were consistent with other studies (Xu et al., 2008; Potter et al., 1996). The major source

10951

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



for atmospheric N₂O locates in southeastern continental North America, including the south part of central Canada, southeastern USA, and all of Mexico, which is associated with cropland distributions and relatively high temperatures (Fig. 3). The weak source in northern Canada is probably due to its low temperature and rainfall, as N₂O emission is significantly controlled by temperature and soil moisture (Chapuis-Lardy et al., 2007; Conrad, 1996; Goldberg and Gebauer, 2009).

N₂O 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 N₂O 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 N₂O emission from terrestrial ecosystems. O₃ has been confirmed to be a pollutant which may decrease productivity (Ashmore, 2005; Wang et al., 2007), and thus suppress carbon source for microbial processes responsible for N₂O production, and finally cause decreases in N₂O emission. However, it is also reasonable to infer that the O₃ pollution inhibits productivity (Wang et al., 2007); and less N uptake might increase N availability in soil and in turn increase N₂O emission. This study showed that the O₃ pollution enhanced N₂O emission from terrestrial ecosystems in southeastern USA due to slightly higher N availability.

The elevated atmospheric CO₂ decreased N₂O emission at continental scale; while the effects varied across North America. The stimulation effects of elevated atmospheric CO₂ on N₂O emission were also observed over southeastern USA and portions of Mexico (Fig. 3c). In field experiments, both positive and negative effects of elevated atmospheric CO₂ on N₂O emission were observed (Kammann et al., 2008; Phillips et al., 2001). The positive or negative effects might be determined by soil N availability, since a field experiment concluded that a small amount of N fertilizer will relieve N limitation under elevated CO₂ concentration (Kettunen et al., 2007). If no progressive N limitation occurs under elevated CO₂, stimulating N₂O emission will be observed,

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

and vice versa. As the theory of progressive N limitation predicts (Luo et al., 2004), rising atmospheric CO₂ could lead to low N availability in soil and thus lead to low N₂O emission (Kettunen et al., 2005; Barnard et al., 2004). In this study, the elevated atmospheric CO₂ substantially decreased the N₂O 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 N₂O flux

The increasing long-term trend and substantial inter-annual fluctuation in terrestrial N₂O flux over North America during 1979–2010 were 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 N₂O over North America, and solely dominated the N₂O flux over Canada. N deposition, O₃ pollution, and atmospheric CO₂ concentrations increased at relatively stable rates through the studied 32 years, which resulted in the long-term trend of N₂O flux (Fig. 8).

Using stepwise regression analysis, we found that climate variability was the predominating factor controlling inter-annual variations in terrestrial N₂O flux at both continental and country levels. Following climate variability, multiple-factor interaction played an essential role in contributing temporal variations in terrestrial N₂O flux. This is consistent with our previous analysis showing that climate variability, followed by multiple-factor interaction, co-dominated inter-annual variations in terrestrial N₂O flux at both continental and country levels (Sect. 3.6). The importance of climate variability in controlling inter-annual N₂O emission indicates the important role of climate variability in controlling variations in atmospheric N₂O concentration (Fluckiger et al., 1999).

BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



4.4 Interactions among multiple factors

The interaction among global change factors has been recognized long before (Der-
mody, 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 multiple-factor interaction was recognized playing
an important role in contributing to terrestrial N₂O flux (Sect. 3.6). This study also re-
vealed that the interactive effects among climate variables predominately control N₂O
emission variation induced by climate variability. Since the various climate variables
are associated in reality, the separation of each climate variable might be biased; the
interaction among climate variables dominates the climate effects. Because the field
experiments are usually labor intensive, multiple-factor experiment might be hard to
carry out; the interactive effects among more than three factors are still short of inves-
tigation (Heimann and Reichstein, 2008; Norby and Luo, 2004). This study shows that
the modeling approach may serve as one complementary tool for field experiments in
addressing interactive effect in a multiple-factor world (Norby and Luo, 2004).

4.5 Uncertainties

This study was able to attribute the spatial and temporal variations in N₂O flux over
North America's terrestrial ecosystems during 1979–2010, but there are several issues
that still need to be improved. Firstly, this study only considered the land conversion
between cropland and natural vegetation, and would have generated more accurate
results if deforestation and afforestation had been included. Secondly, the pre-1979
legacy effect may cause some biases in this study, which solely analyzed N₂O flux
over the time period of 1979–2010. Thirdly, the soil may act as a sink of atmospheric
N₂O (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 N₂O flux at both continental and county levels. More
field studies are needed to understand the mechanisms for N₂O sink in soil, which will

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



improve the regional estimation of N₂O flux. Fourthly, although we have compared our estimated factorial impacts on N₂O flux with other studies, the effects of global change factors on N₂O have not been comprehensively calibrated and validated because of the scarcity of field observations on factorial N₂O fluxes (Dermody, 2006). Fifthly, the thawing-freezing cycle has long been identified as a major factor on N₂O emission, especially in temperate and boreal regions (de Bruijin et al., 2009; Repo et al., 2009; Christensen and Tiedje, 1990), yet the real mechanisms are still not well investigated (de Bruijin et al., 2009). So this study might have underestimated the N₂O emission due to shortage of the effects of freezing-thawing. Sixthly, although the regional data used in this study are fairly current and 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 model associated with more field observations might provide more accurate nitrogen deposition data. Last but not least, it will be an improvement when modeling studies can take into account more environmental factors such as wild and prescribed fires, harvests, insect outbreaks, etc.

5 Conclusions

This study examined the factorial contributions to the terrestrial N₂O 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 CO₂, O₃ pollution, N fertilizer application, and land conversion. 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 N₂O concentration as well as beneficial for the policy-making for curbing the increase in atmospheric N₂O concentration. The complicated effects of multiple-factor interaction on N₂O flux suggest that the current field experiments, which usually ignore the interactive effects from multiple-factors, may

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



lead to biases in the estimation of N₂O flux. This study also pointed out that the models driven by only a few global change factors may bring bias in estimating N₂O flux.

This study is among the first attempts to attribute the spatiotemporal variations in regional terrestrial N₂O flux to multiple global change factors over a long period of time. A collaborative effort between field ecologists and modelers is necessary for further investigation of the underlying mechanisms for spatial and temporal variations in N₂O flux.

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BGD

8, 10935–10977, 2011

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Controls on
terrestrial N₂O flux in
North America**

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. Experimental design for this study.

Simulation	Climate	Nitrogen deposition	CO ₂	O ₃	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
	Climate (precipitation)	Climate (maximum, average, minimum temperature)	Solar radiation	Relative humidity	Nitrogen deposition, CO ₂ , O ₃ , nitrogen fertilizer, and land conversion	
15	1900–2010	1900–1979	1900–1979	1900–1979		1900
16	1900–1979	1900–2010	1900–1979	1900–1979		1900
17	1900–1979	1900–1979	1900–2010	1900–1979		1900
18	1900–1979	1900–1979	1900–1979	1900–2010		1900
19	1900–1979	1900–1979	1900–1979	1900–1979		1900

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

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 2. Changing rates of driving factors for DLEM simulations.

	Variables	Changing rates (mean ± SD)
Climate	Maximum temperature (°C a ⁻¹)	0.04 ± 0.01*
	Minimum temperature (°C a ⁻¹)	0.03 ± 0.01*
	Average temperature (°C a ⁻¹)	0.03 ± 0.01*
	Precipitation (mm a ⁻¹)	0.09 ± 0.62
	Relative humidity (% a ⁻¹)	-0.01 ± 0.01
	Solar radiation (W m ⁻² a ⁻¹)	0.19 ± 0.03*
Others	O ₃ pollution (ppm-h a ⁻¹)	0.96 ± 0.08*
	N deposition (mg m ⁻² a ⁻¹)	1.96 ± 0.05*
	N fertilizer application (mg m ⁻² a ⁻¹)	0.02 ± 0.01*
	Atmospheric CO ₂ concentration (ppm a ⁻¹)	1.68 ± 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.

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 ²)	4.05	6.93 ~ 6.99	3.57 ~ 3.59	2.61 ~ 2.64	2.06 ~ 2.07	0.53 ~ 0.60	2.51 ~ 2.59
Percentage	18.09	31.10	15.98	11.72	9.23	2.49	11.39

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

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Table 4. Factorial contributions to the accumulated N₂O from 1979 to 2010 (Combined represents the effects with all six factors being considered; “Climate” represents the impacts of climate variability only; “Ndep” represents the impacts of N deposition; “CO₂” represents the impacts of CO₂ variation; “O₃” represents the impacts of O₃ pollution; “Nfer” represents the impacts of N fertilizer application; “Land conversion” represents the impacts of land cover change only; “Interaction” represents the balance of all interactive effects of the six environmental factors).

		Baseline	Climate	Ndep	CO ₂	O ₃	Nfer	Land conversion	Interaction	Total flux
USA	Accumulative N ₂ O flux (TgN)	32.22±0.44	-0.47±0.57	0.23±0.02	-0.23±0.03	0.09±0.02	0.96±0.10	0.25±0.02	0.44±0.29	33.49±0.57
	Percentage (%)	96.22	-1.41	0.69	-0.68	0.27	2.86	0.76	1.30	100
Canada	Accumulative N ₂ O flux (TgN)	8.85±0.12	0.75±0.33	0.03±0.005	-0.21±0.02	0.004±0.0007	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	Accumulative N ₂ O flux (TgN)	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	Accumulative N ₂ O flux (TgN)	52.32±0.57	0.32±0.70	0.49±0.06	-0.52±0.07	0.11±0.02	1.21±0.12	0.21±0.02	0.95±0.33	55.08±0.84
	Percentage (%)	94.99	0.59	0.88	-0.95	0.20	2.20	0.38	1.72	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 N₂O flux; negative values represent inhibiting effects of global change factors on N₂O flux.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Table 5. Contributions from individual climate variable to the climate-induced N₂O accumulation (Tg N) from 1979 to 2010.

	Precipitation	Relative humidity	Solar radiation	Temperature	Interaction	Total flux
USA	-3.09 ± 0.48	-4.01 ± 0.36	-3.90 ± 0.35	-0.35 ± 0.43	10.89 ± 1.06	-047 ± 0.57
Canada	0.19 ± 0.27	-1.67 ± 0.16	-1.75 ± 0.15	-1.28 ± 0.17	5.27 ± 0.46	0.75 ± 0.33
Mexico	-0.29 ± 0.42	-0.70 ± 0.32	-0.71 ± 0.32	0.69 ± 0.38	1.06 ± 0.97	0.05 ± 0.41
North America	-3.19 ± 0.65	-6.38 ± 0.51	-6.36 ± 0.51	-0.95 ± 0.65	17.08 ± 1.56	0.32 ± 0.70

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 climate variables on N₂O flux; negative values represent inhibiting effects of climate variables on N₂O flux.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

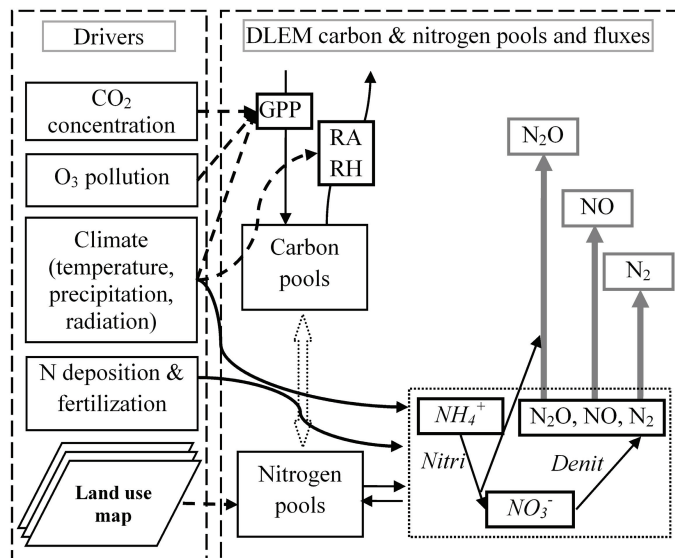
Interactive Discussion

Table 6. Comparison of factorial effects on N₂O fluxes against other studies (positive values mean increase; negative values mean decrease in N₂O emission).

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

Controls on terrestrial N₂O flux in North America

X. F. Xu et al.



Major processes: *Nitr*: 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 N₂O processes.

Fig. 1. Conceptual diagram showing major processes for N₂O flux in response to multiple global change factors in the DLEM model.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



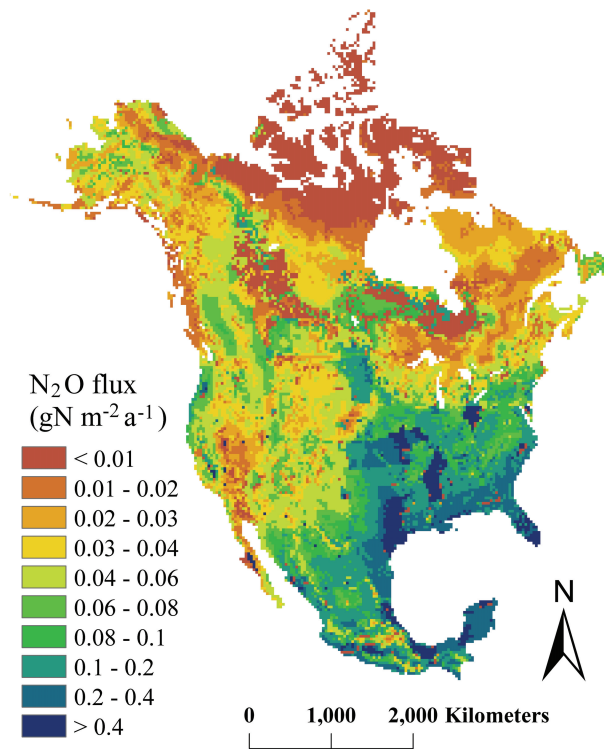


Fig. 2. Spatial variations of terrestrial N₂O fluxes caused by global change factors over North America from 1979 to 2010.

**Controls on
terrestrial N₂O flux in
North America**

X. F. Xu et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

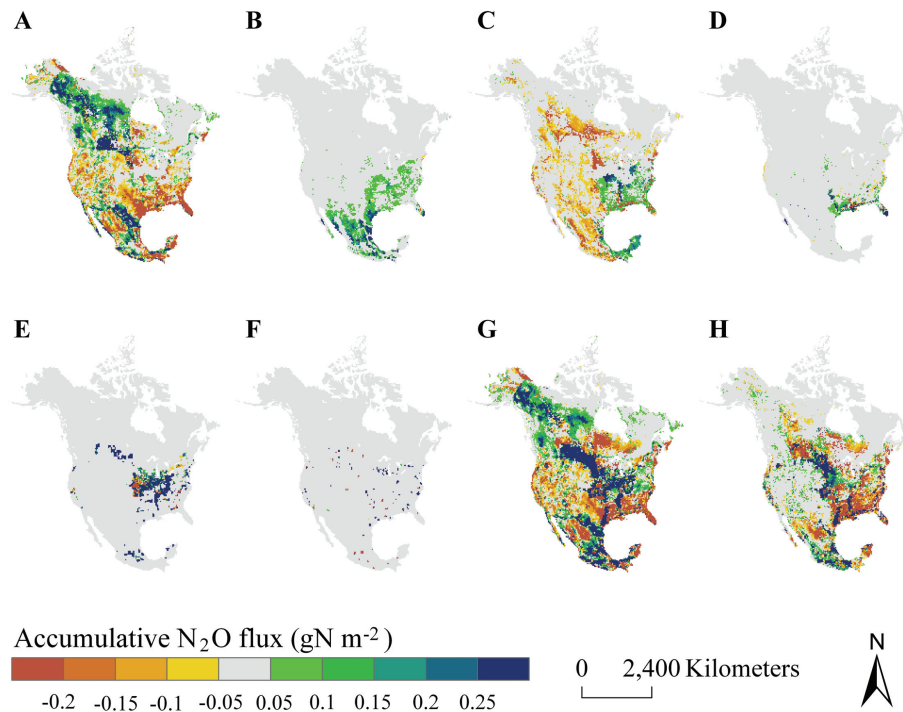


Fig. 3. Factorial contributions to the spatial variations in accumulated N₂O flux over North America from 1979 to 2010 ((**A**) climatic variability; (**B**) N deposition; (**C**) CO₂; (**D**) O₃ pollution; (**E**) N fertilizer application; (**F**) land conversion; (**G**) all combined; (**H**): interaction).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



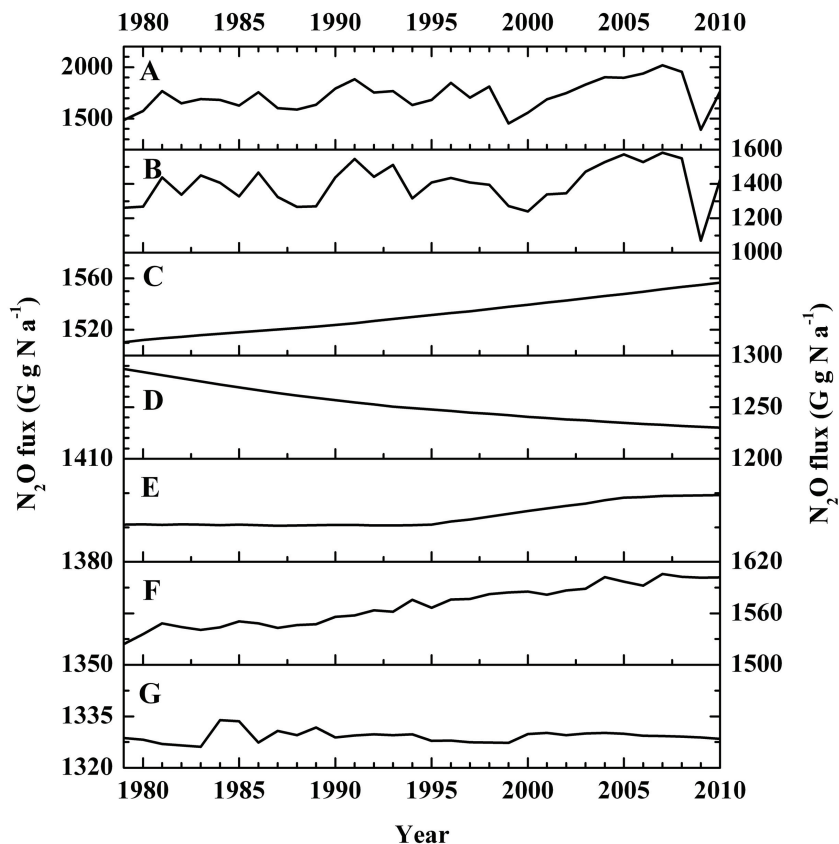


Fig. 4. Temporal variations of terrestrial N_2O flux caused by global change factors over North America from 1979 to 2010 ((**A**) all combined simulation; (**B**) climate only simulation; (**C**) N deposition only simulation; (**D**) CO_2 only simulation; (**E**) O_3 pollution only simulation; (**F**) N fertilizer application only simulation; (**G**) land conversion only simulation).

**Controls on
terrestrial N_2O flux in
North America**

X. F. Xu et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



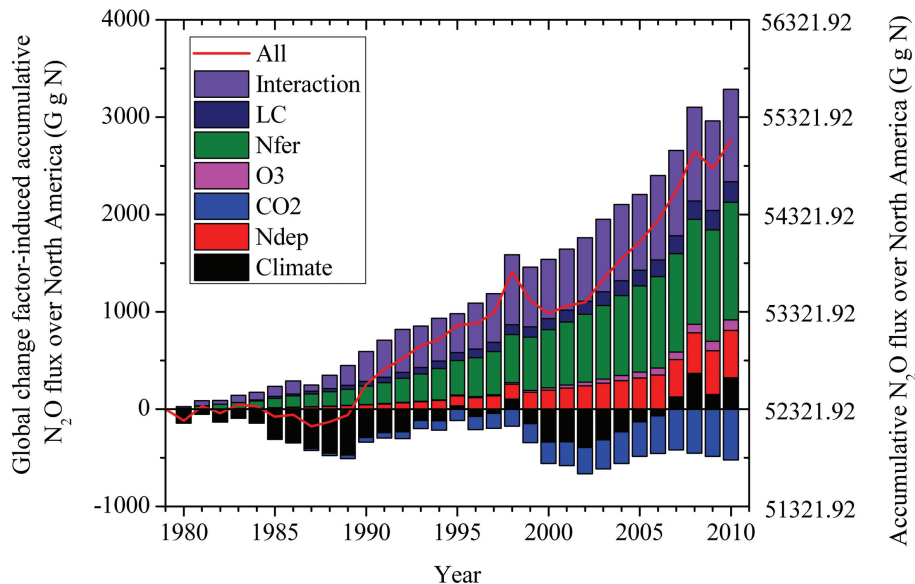


Fig. 5. Factorial contributions to accumulated N_2O flux over North America during 1979–2010 (The right Y-axis shows the accumulative N_2O flux with baseline; “All” means N_2O flux derived from all combined simulation; “Interaction” means contribution from multiple-factor interaction; “LC” means contribution from land conversion; “Nfer” means contribution from N fertilizer application; “O₃” means contribution from O₃ pollution; “CO₂” means contribution from elevated atmospheric CO₂; “Ndep” means contribution from N deposition; “Climate” means contribution from climate variability).

Controls on terrestrial N_2O flux in North America

X. F. Xu et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

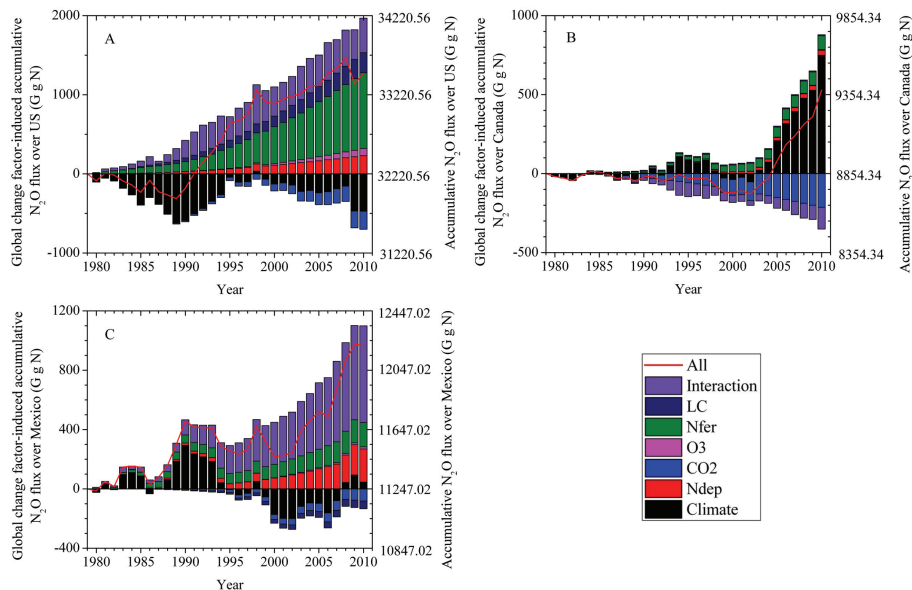


Fig. 6. Factorial contributions to accumulated N₂O flux at country-level during 1979–2010 ((**A**) United States of America; (**B**) Canada; (**C**) Mexico) (The right Y-axis shows the accumulative N₂O flux with baseline; “All” means N₂O flux derived from all combined simulation; “Interaction” means contribution from multiple-factor interaction; “LC” means contribution from land conversion; “Nfer” means contribution from N fertilizer application; “O₃” means contribution from O₃ pollution; “CO₂” means contribution from elevated atmospheric CO₂; “Ndep” means contribution from N deposition; “Climate” means contribution from climate variability).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Controls on terrestrial N₂O flux in North America

X. F. Xu et al.

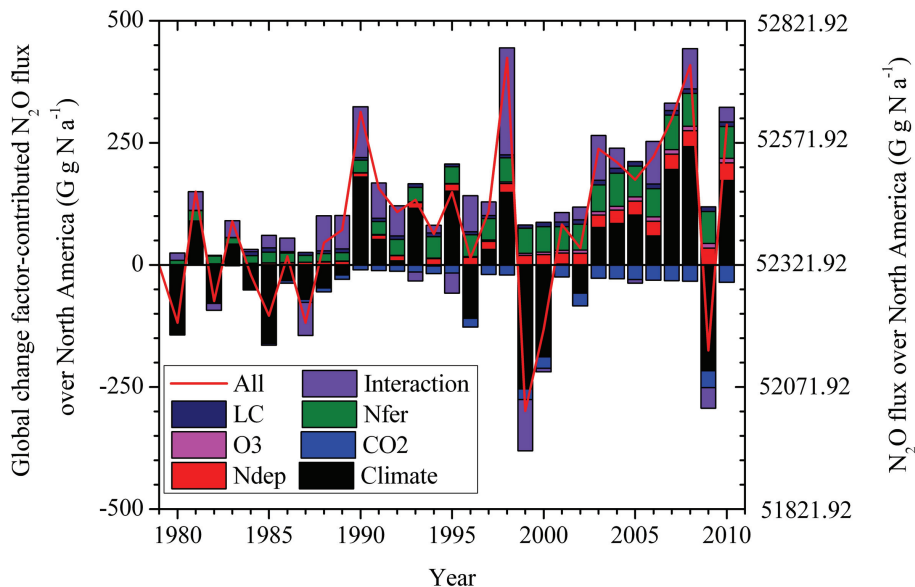


Fig. 7. Factorial contributions to the inter-annual variations in N₂O flux over North America (The right Y-axis shows the N₂O flux with baseline; “All” means N₂O flux derived from all combined simulation; “Interaction” means contribution from multiple-factor interaction; “LC” means contribution from land conversion; “Nfer” means contribution from N fertilizer application; “O₃” means contribution from O₃ pollution; “CO₂” means contribution from elevated atmospheric CO₂; “Ndep” means contribution from N deposition; “Climate” means contribution from climate variability).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



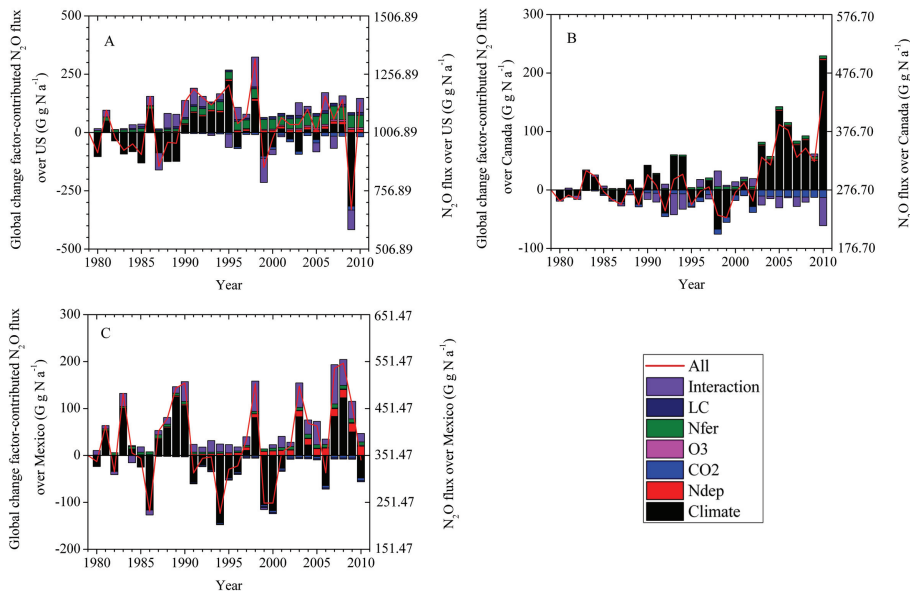


Fig. 8. Factorial contributions to the inter-annual variations in N_2O flux at country level ((**A**) United States of America; (**B**) Canada; (**C**) Mexico) (The right Y-axis shows the N_2O flux with baseline; “All” means N_2O flux derived from all combined simulation; “Interaction” means contribution from multiple-factor interaction; “LC” means contribution from land conversion; “Nfer” means contribution from N fertilizer application; “ O_3 ” means contribution from O_3 pollution; “ CO_2 ” means contribution from elevated atmospheric CO_2 ; “Ndep” means contribution from N deposition; “Climate” means contribution from climate variability).

Controls on terrestrial N_2O flux in North America

X. F. Xu et al.

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

