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Chaoqun (Crystal) Lu, *Auburn University Main Campus*
Hanqin Tian, *Auburn University Main Campus*
Mingliang Liu, *Auburn University Main Campus*
Wei Ren, *Auburn University Main Campus*
Xiaofeng Xu, *Auburn University Main Campus*, et al.

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Effect of nitrogen deposition on China's terrestrial carbon uptake in the context of multifactor environmental changes

Chaoqun Lu,[1,2] Hanqin Tian,[1,2,4] Mingliang Liu,[1,2] Wei Ren,[1,2] Xiaofeng Xu,[1,2] Guangsheng Chen,[1,2] and Chi Zhang[1,3]

[1]*Ecosystem Dynamics and Global Ecology Laboratory, School of Forestry and Wildlife Sciences, Auburn University, Auburn, Alabama 36849 USA*
[2]*International Center for Climate and Global Change Research, Auburn University, Auburn, Alabama 36849 USA*
[3]*Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011 China*

Abstract. The amount of atmospheric nitrogen (N) deposited on the land surface has increased globally and by nearly five times in China from 1901 to 2005. Little is known about how elevated reactive N input has affected the carbon (C) sequestration capability of China's terrestrial ecosystems, largely due to the lack of reliable data on N deposition. Here we have used a newly developed data set of historical N deposition at a spatial resolution of 10 km × 10 km in combination with other gridded historical information on climate, atmospheric composition, land use, and land management practices to drive a process-based ecosystem model, the dynamic land ecosystem model (DLEM) for examining how increasing N deposition and its interactions with other environmental changes have affected C fluxes and storage in China's terrestrial ecosystems during 1901–2005. Our model simulations indicate that increased N deposition has resulted in a net C sink of 62 Tg C/yr (1 Tg = 10^{12} g) in China's terrestrial ecosystems, totaling up to 6.51 Pg C (1 Pg = 10^{15} g) in the past 105 years. During the study period, the N-induced C sequestration can compensate for more than 25% of fossil-fuel CO_2 emission from China. The largest C sink was found in southeast China, a region that experienced the most significant increase of N deposition in the period 1901–2005. However, the net primary productivity induced by per-unit N deposition (referred to as ecosystem N use efficiency, ENUE, in this paper) has leveled off or declined since the 1980s. This indicates that part of the deposited N may not be invested to stimulate plant growth, but instead leave the ecosystem by various pathways. Except shrubland and northwest/southwest China, signs of N saturation are apparent in the rest major biome types and regions, with ENUE peaking in the 1980s and leveling off or declining thereafter. Therefore, to minimize the excessive N pollution while keeping the N-stimulated C uptake in China's terrestrial ecosystems, optimized management practices should be taken to increase N use efficiency rather than to keep raising N input level in the near future.

Key words: carbon dynamics; China; dynamic land ecosystem model; nitrogen deposition; terrestrial ecosystem.

Introduction

Anthropogenic activities have substantially altered the pathway and magnitude of nitrogen (N) flows from the atmosphere to terrestrial ecosystems, which has consequently enhanced the reactive N amount at a record pace (Vitousek et al. 1997, Galloway et al. 2004, 2008*b*). At the global scale, N deposited onto the land surface is unevenly distributed and concentrated in intensively cultivated and industrialized regions (Townsend et al. 1996, Dentener 2006). Among them, the northeastern United States, western and central Europe, and eastern Asia have attracted much attention for their exposure to remarkably elevated N deposition (Holland et al. 2005, Galloway et al. 2008*a*). The atmospheric transport model indicates that N deposited on land surface of China summed up to 0.93 Tg N in the 1860, accounting for 3% of global N deposition, while it amounted to 8.8 Tg N in the early 1990s, around 9% of the global N deposition input (Dentener 2006, Galloway et al. 2004). Nonetheless, the model-derived atmospheric N input data set commonly used for driving ecosystem models was shown to largely underestimate the actual N deposition rate in China, by nearly 35% less than monitoring values (Lu and Tian 2007). Our previous work pointed out that current N deposition rates in China are close to or even exceed those in Europe and the United States (Lu and Tian 2007). Due to population increase, food demands, and industrial development, the emission and deposition of reactive N are projected to continue to increase in China over the coming decades (Richter et al. 2005, Dentener 2006, Galloway et al. 2008*a*). Atmospheric N enrichment and the consequent alterations in ecosystem functioning

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[4] Corresponding author. E-mail: tianhan@auburn.edu

across China's terrestrial ecosystems have attracted increasing attention from both the scientific community and policy makers in recent years (Tian et al. 2006, Lu et al. 2007). Especially in some areas of southeast China, the N deposition rate reaches up to more than 40 kg $N \cdot ha^{-1} \cdot yr^{-1}$ due to the large amount of fossil fuel combustion and fertilizer application (e.g., wet deposition of 50.8 kg $N \cdot ha^{-1} \cdot yr^{-1}$ in Hangzhou as measured by Shui et al. [1999]; wet deposition of dissolved inorganic and organic N at 32–34 and 18 kg $N \cdot ha^{-1} \cdot yr^{-1}$, respectively, in Dinghushan measured by Fang et al. [2008]). However, the impacts of increased N deposition on carbon (C) sequestration capacity in China's terrestrial ecosystems still remain far from certain, since none of large-scale assessment has been performed in this region although numerous studies have investigated N deposition and its related C budget on a global scale (e.g., Churkina et al. 2007, Xu-Ri and Prentice 2008, Jain et al. 2009, Zaehle et al. 2010*b*). Given the critical role of N deposition in mitigating climate change, it is of particular importance to accurately estimate the N-induced C dynamics in China, as one of major components in determining greenhouse gas emission/uptake.

Previous studies found that the N fertilization effect is partly responsible for the "missing C sink" (Townsend et al. 1996, Holland et al. 1997, Reich et al. 2006*a*, Magnani et al. 2007). Especially for N-limited ecosystems such as temperate forests, anthropogenic N deposition plays a critical role in stimulating plant growth, altering soil respiration, and determining C allocation and the consequent C storage pattern on the Earth (Vitousek and Howarth 1991). However, field studies from Europe and the United States have documented that prolonged inputs of 10 to 20 kg $N \cdot ha^{-1} \cdot yr^{-1}$ have led to nitrogen saturation (Fisher et al. 2007). The potentially detrimental effects of excessive N have been as well highlighted in non-N-limited or N-saturated systems (Aber et al. 1998, Matson et al. 1999, Asner et al. 2001, Fenn et al. 2003). The nonlinear responses make the estimation of N-induced C sink more complicated.

A wide range of N-addition experiments and N-gradient experiments, most in the short term, have been carried out to examine the changes of plant growth, soil respiration, and N retention and loss in response to chronic N loading or N removal in various ecosystems (Wright and Rasmussen 1998, Bowden et al. 2004, Magill et al. 2004, Mo et al. 2004, Fang et al. 2008, Niu et al. 2009). These studies provide insightful points on the effects of altered N availability although the reported ecosystem responses are divergent, or even controversial due to heterogeneous local conditions. In addition, it's also difficult to identify N effects while taking into account its interaction with other environmental changes. Due to the complexity of the C–N interaction, the heterogeneity of land ecosystems and the limitation of financial availability, it is nearly impossible to conduct large-scale field experiments especially in a long time period. Spatially explicit process-based ecosystem models, in contrast, have the capability to distinguish the large-scale long-term impacts of individual environmental change through conducting factorial simulation experiments (Tian et al. 2011*b*). Hence, the ecosystem models that simulate the fundamental mechanisms controlling C and N cycles in terrestrial ecosystems can be used as effective tools to investigate the N-induced C dynamics in the context of multiple environmental stresses at large scale.

By using the dynamic land ecosystem model (DLEM; Tian et al. 2011*b*, *c*), driven by spatially explicit data of N deposition rate and other environmental changes including rising atmospheric carbon dioxide (CO_2) and ozone (O_3), increasing fertilizer uses and changing climate and land use, in this study, we intend to examine how increasing N deposition and other environmental stresses have interactively affected the C sequestration capacity of China's terrestrial ecosystem during 1901–2005. We select this time period because it covers the major turning points and increasing periods of anthropogenic N deposition in China since the preindustrial era. Specifically, we have tried to address the following questions: (1) To what extent had N deposition influenced C storage in China over the past 105 years? (2) How did elevated N deposition alter terrestrial C sequestration capacity in spatial and temporal contexts? (3) How did different biomes respond to increased N input across China? (4) What gaps in knowledge, data and model are causing the uncertainties in estimating N deposition effects on the C cycle?

Methods and Model Strategy

Model description

The dynamic land ecosystem model (DLEM) is a highly integrated process-based model which aims at simulating the interactions and feedbacks among/within the terrestrial ecosystem components in the face of multiple natural and anthropogenic disturbances, such as changes in climate, atmospheric compositions (CO_2, O_3, reactive N), land use/cover patterns, land management practices (harvest, rotation, fertilization, irrigation, etc.), and natural perturbations (fire, insect/disease, hurricane, etc.). DLEM involves fully coupled C–N–water cycles to estimate the hydrological and biogeochemical fluxes and pool sizes at multiple scales in time from daily to yearly and space from site to region/globe. Up to this point, DLEM has been widely applied and well documented in the studies highlighting the contribution of multiple stresses to the terrestrial C, N, and water cycles in several typical regions across the globe (e.g., Chen et al. 2006, Ren et al. 2007, Liu et al. 2008, Zhang et al. 2007, 2010, Tian et al. 2008, 2010*a*, *d*, 2011*a*, *b*, *c*).

In this study, we adopted net carbon exchange (NCE), and C storage change in vegetation, soil, and litter pools as major indices to investigate the ecosystem

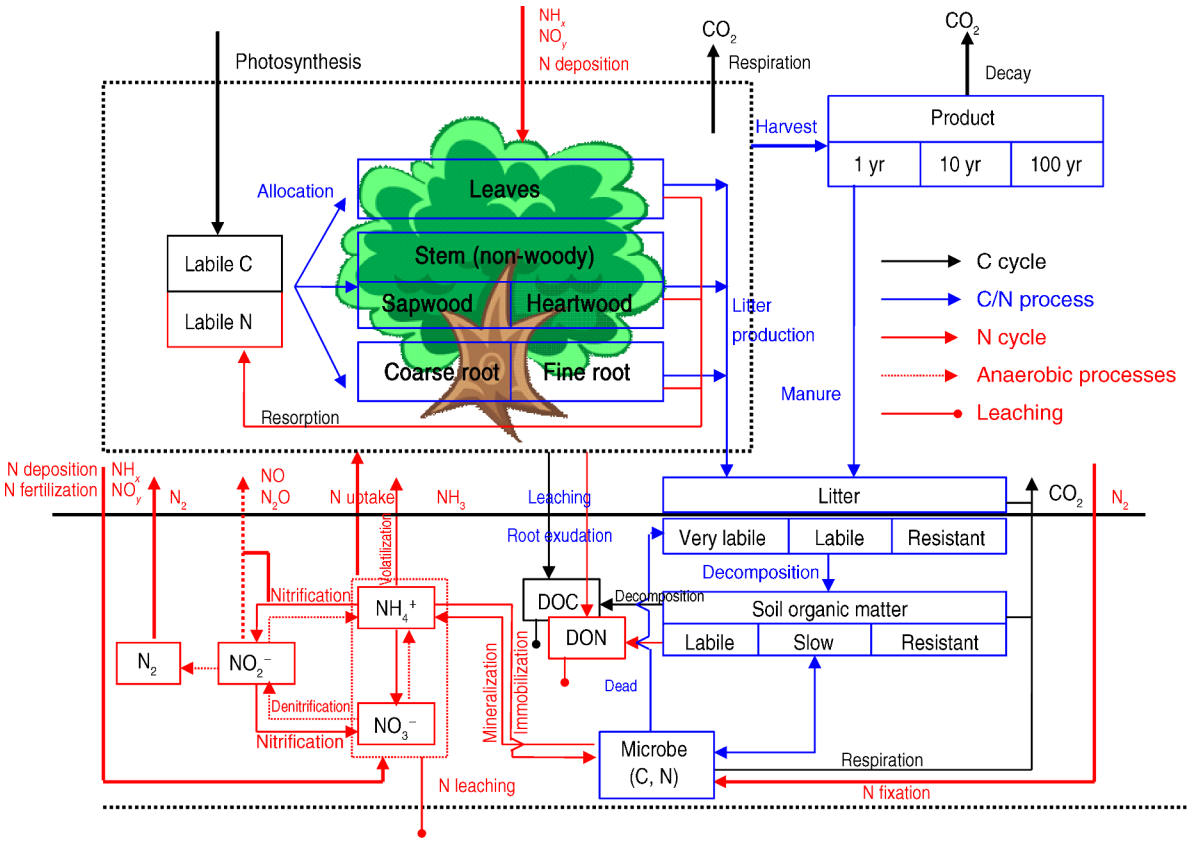

FIG. 1. Nitrogen processes and C–N coupling in the dynamic land ecosystem model (DLEM).

C dynamics in response to atmospheric N input in China. The model output of NCE is calculated as the difference between C assimilation through photosynthesis (gross primary production, GPP) and C loss to the atmosphere (Eq. 1), equivalent to the net ecosystem carbon balance (NECB) defined in Tian et al. (2011*b*):

$$\mathrm{NCE} = \mathrm{GPP} - R_a - R_h - E_c - E_p - \mathrm{CH_4} \qquad (1)$$

where R_a is autotrophic respiration, R_h is heterotrophic respiration, E_c is the C flux associated with land conversion, E_p is C release during the consumption of agricultural and wood products; and CH_4 is the net CH_4 exchange between terrestrial ecosystems and the atmosphere. The positive and negative values of NCE indicate carbon uptake from and carbon release to the atmosphere, respectively.

DLEM partitions C and N cycles into vegetation, litter, soil, microbe, and product components (Fig. 1). Vegetation pool is composed of six boxes for woody species (storage, leaf, heartwood, sapwood, fine root, and coarse root) and five boxes for herbaceous species (storage, leaf, stem, fine root, and coarse root). Both aboveground and belowground litter pools include three boxes with different turnover rates (resistant, labile, very labile). Soil organic C and N are partitioned into labile, slow, and resistant pools, while soil inorganic N pool includes NO_3^--N and NH_4^+-N. The product pool is divided into three boxes with 1-, 10- and 100-year lifetimes. In DLEM, N processes are intimately coupled to the C processes by different biomass compartment-specific C:N ratios. When the amount of storage N is not enough to maintain the plant C:N ratio, the plant growth will be limited by available N, and excessive C will be stored in the C storage pool. Likewise, when plant growth is limited by C, excessive N will be stored in the N storage pool. DLEM has prescribed the minimum and maximum load for storage pool and both C and N storage are involved in mass allocation among tissues. DLEM simulates the N in several forms: organic N stored in biomass, labile N stored in plants, organic N stored in soil and litter/woody debris, soil dissolved organic N (DON), and soil inorganic N ions such as NH_4^+ or NO_3^-.

In DLEM, the N cycle is fully open, i.e., N inside the ecosystem can be exchanged with external sources and sinks through deposition, leaching, nitrous gas emissions, and so on. Combined with the inside mechanisms such as plant uptake and N mineralization, the open-N cycle provides a buffer to control the fluctuation of interior N (Rastetter et al. 1997). N flows follow mass balance principle, where change of ecosystem available N depends on the difference between N inputs (e.g., N deposition, fertilization, fixation) and N outputs (e.g., N

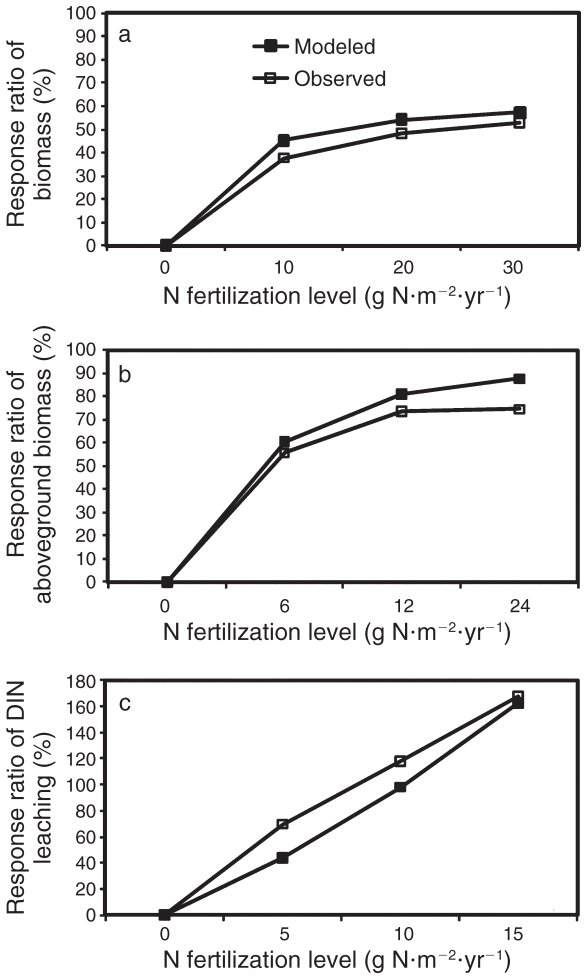

FIG. 2. Comparison of (a, b) modeled biomass increment and (c) dissolved inorganic nitrogen (DIN) leaching loss in response to different doses of N addition with manipulated N fertilization experiments on (a) maize crop of Yucheng, China, in 2001 (Wang et al. 2005); (b) natural wetland of Sanjiang, China, in 2008 (C. C. Song, L. L. Wang, H. Q. Tian, D. Y. Liu, Z. M. Wan, C. Q. Lu, L. H. Zhang, Y. C. Li, and R. J. Ge, *unpublished manuscript*); and (c) subtropical evergreen broad-leaved forest of Dinghushan, China, in 2004 (Fang et al. 2009). The response ratio is calculated as $R = ([V_{\mathrm{Nfer}} - V_{\mathrm{ctrl}}]/V_{\mathrm{ctrl}})$, where V_{Nfer} and V_{ctrl} are biomass or leached DIN amount with and without N fertilizer applied, respectively.

leaching, N_2O, NO emission, NH_3 volatilization). Since detailed information on DLEM is accessible in other published papers (Ren et al. 2007, 2011*a*, *b*, Zhang et al. 2007, Liu et al. 2008, Tian et al. 2010*a*, *c*, *d*, 2011*a*, *b*, *c*, Xu et al. 2010), we primarily introduce the model strategy related to N budget and N control on C cycling in this study (details can be found in the Appendix).

Model calibration and validation

Before model simulation, we calibrated DLEM to get fitting parameter values for each plant functional type by minimizing the differences between the simulated C, N, water fluxes, and site-level observations. Field measurements from 11 long-term experiment stations in Chinese Ecological Research Network (CERN) and other sites reported by published paper, covering all the plant functional types in our study, have been selected to calibrate the key parameters of DLEM (Ren et al. 2011*a*, *b*, Tian et al. 2011*b*, c). Observations of fluxes of CO_2, CH_4, N_2O, evapotransporation (ET), GPP, net primary production (NPP), C, and N content in vegetation and soil pools, N uptake, N leaching, and so on are used to characterize each site.

For model simulation of China's terrestrial ecosystems, DLEM has been well validated against site-level CO_2 flux measurements, national and biome-level inventories of C fluxes and pools, atmospheric inversions of CO_2 balance, and remote sensing-derived GPP/NPP. It has been shown that DLEM performs well in capturing the magnitude and spatial and temporal (interannual, seasonal, daily) patterns of net C exchange (Tian et al. 2011*b*), balance of terrestrial CO_2, CH_4, and N_2O (Tian et al. 2011*c*), and water fluxes in China (Liu et al. 2008) and the sub-continent including China, e.g., monsoon Asia (Tian et al. 2011*a*). In addition, DLEM has been intensively validated against observation data outside of China, including site-level observed daily and seasonal patterns of NEE (Schwalm et al. 2010), CH_4 and N_2O fluxes (Tian et al. 2010*d*), and field observed GPP and ET at daily and annual time step (Tian et al. 2010*a*) in North America.

Instead of repeatedly evaluating DLEM's capability in simulating C and water balance in China's terrestrial ecosystems over a certain time period, therefore, we would highlight the validation of simulated ecosystem dynamics in response to factorial variation in this study through the following two ways. First, the modeled response of plant growth to N addition has been validated against the manipulated experiments conducted on maize crop in northern China and natural wetland in northeastern China, where three levels of N fertilizer were applied in the sampling plots (Fig. 2a and b). Our simulated response agreed well with the observed responses of the experiments, with the low N fertilizer level (10 g $N \cdot m^{-2} \cdot yr^{-1}$ in cropland and 6 g $N \cdot m^{-2} \cdot yr^{-1}$ in wetlands) resulting in the largest increase in biomass while additional N input yielding less increase in both observed and modeled biomass. The modeled response of dissolved inorganic nitrogen (DIN) leaching has also been verified against the observations with three doses of N fertilizers in a subtropical monsoon forest in southern China (Fig. 2c). The field experiments showed that DIN leaching loss continued to increase and the response ratio was up to over 160% in the high level of N addition (15 g $N \cdot m^{-2} \cdot yr^{-1}$) relative to the control. The DLEM model can well capture the overall trajectory of leaching loss in response to N fertilization. However, the modeled response was 20% lower than observation when low and medium levels of N fertilizer were applied. That is because DLEM oversimplifies the dynamics of stream flow in the pulse of heavy rainfall, which underestimated

TABLE 1. The simulation experiments for evaluating modeled response of plant growth to changes in atmospheric N deposition rate and CO_2 concentration.

Experiment	Climate	CO_2 (ppm)	O_3	Ndep	Nfer	LUCC
Low N (N_a)	1900	296	excluded	2.5 kg N $ha^{-1}yr^{-1}$	1900	1900
High N (N_e)	1900	296	excluded	250 kg N $ha^{-1}yr^{-1}$	1900	1900
Low CO_2(C_a)	1900	350	excluded	1993†	1900	1900
High CO_2(C_e)	1900	550	excluded	1993	1900	1900

Notes: The subscript "a" indicates ambient level, and "e" indicates elevated level. Ndep, Nfer, and LUCC are abbreviation for N deposition, N fertilization, and land use and cover change, respectively. Dates indicate that environmental factors in those time periods were used to drive the model.

† We used preindustrial CO_2 level when checking the N addition effect; N deposition of the year 1993 was used to reduce the N limitation in checking CO_2 fertilization effect.

runoff and thus DIN leaching losses, especially when soil N content is relatively low. Second, randomly selected sites were used to examine the simulated NPP change induced by N and CO_2 enrichment. Our previous work shows that increases in atmospheric N deposition and CO_2 concentration are among the greatest contributors to China's terrestrial C sink during 1961–2005 (Tian et al. 2011*b*). Hence, we conduct a series of model simulations to further evaluate DLEM's capability in attributing C sink to N addition and CO_2 enrichment. In order to get the responses of modeled plant growth to elevated N input and atmospheric CO_2 concentration, we set up four simulation experiments with different N and CO_2 input levels (N addition, 2.5–250 kg N·ha^{-1}·yr^{-1}; CO_2, 350–550 ppm; Table 1) while keeping climate, N fertilization, and land use patterns at the level of 1900 (data details can be found in *Input data*), and randomly select 10 grid cells for each vegetation type. The model is fed with these driving forces to reach equilibrium and then we calculate the response ratio of interested variables in elevated and control experiments. It demonstrates that the modeled responses of plant growth to different doses of N addition and CO_2 concentration fall within the ranges of field experiments/observations (Table 2) which are claimed by meta-analysis with multiple studies (Ainsworth and Long 2005, LeBauer and Treseder 2008).

Input data

N deposition data.—Although N deposition data from three-dimensional chemical transport models are commonly used to evaluate the N effects on terrestrial ecosystems (Townsend et al. 1996, Holland et al. 1997, Churkina et al. 2007, Jain et al. 2009), our previous work suggests that the actual deposition rates in China, especially for aqueous NH_4^+, have been significantly underestimated by models (Lu and Tian 2007). In this study, we established an empirical relationship among gridded N deposition data in China during 1992–1993 from the chemical transport model (Dentener 2006), the quarterly average values of aqueous NO_3^- and NH_4^+ concentration collected from 81 rural and remote sites in the same period, and gridded annual average precipitation amounts with a resolution of 10 km × 10 km. Based on this empirical equation, modeled N deposition rate during 1901–2005 has been upregulated to a level closer to field observations. The data from national monitoring network in China is only limited to precipitation chemistry, while Dentener's modeling results include the bulk deposition of NH_x and NO_y. Therefore, we adopted a fixed ratio of 0.7, which is derived from the average of monitoring data and literature, to represent the fraction of wet deposition to the bulk deposition, assuming the spatial heterogeneity of this ratio is negligible. We used a randomly distributed site ensemble to sample the dry N deposition rates from these three different data sources. It turned out that dry deposition rates derived from our approach are much closer to the observations than Dentener's data set (Table 3). The relatively higher correlation coefficient indicated our data could better explain the spatial variation of dry deposition in China. We only include dry deposition of NO_2 in this study since other species such as gaseous HNO_3, gaseous NH_3, particulate NO_3^-, particulate

TABLE 2. Comparison of the simulated and observed responses of primary productivity to N addition and atmospheric CO_2 increase among various biomes.

Biome	Modeled response (%)	Measured response (%)
NPP response to N addition (2.5–250 kg N·ha^{-1}·yr^{-1})†		
Temperate forest	15–27 (21)	11–28 (19)
Tropical forest	33.6–92.8 (63.2)	30–97 (60)
Temperate grass	21.5–63.1 (42.3)	37–71 (53)
Desert	0.4–37.7 (19.1)	−20–55 (11)
Wetland	13–27.1 (20.1)	0–34 (16)
Photosynthesis (GPP) response to CO_2 increase (350–550 ppm)‡		
Forest	27.9–37.1 (32.5)	17–40 (29)
Shrubland	25.8–43.6 (34.7)	33–60 (46)
Grassland	24.9–49.6 (37.3)	28–47 (37)
Crop, wheat§	18.8–27.8 (23.2)	−2–33 (14)
Crop, rice§	11.5–17.7 (14.6)	−7–30 (10)

Note: The response ratio is the ratio of difference in interested variables between elevated and control experiment to the control output. The reported range is the 95% confidence interval. The mean response is shown in parentheses.

† Measured response comes from LeBauer and Treseder (2008).

‡ Measured response comes from Ainsworth and Long (2005).

§ The response ratio is measured on crop yield.

TABLE 3. Comparisons of dry deposition in the years 1993 and 1999 (kg N·ha^{-1}·yr^{-1}) extracted from our data and Dentener's modeled data (Dentener 2006) with the field observations.

			This study		Dentener (2006)	
Year	*N*	Observations	Mean ± SD	*R*	Mean ± SD	*R*
1993	179	2.76 ± 1.34	2.77 ± 0.92	0.712	0.71 ± 0.63	0.382
1999	179	2.80 ± 1.03	2.77 ± 0.98	0.668	0.78 ± 0.73	0.382

Notes: Mean and SD are average and standard deviation of the dry N deposition rate from 179 randomly distributed sites, respectively. *R* indicates the correlation coefficient of our data or Dentener's data to the observations.

NH_4^+, and organic nitrogen are seldomly measured on the long-term monitoring sites.

To demonstrate the spatial pattern of N deposition changes from the years 1901 through 2005, we averaged the N deposition rate in each grid during the first decade (1901–1910) and the last decade (1996–2005). The differences between the first decade and the last decade can indicate the spatial heterogeneity of N deposition increase over the past 105 years, avoiding extreme values in a specific year. It shows that all the areas in China have been experiencing an enhancement of ambient N deposition, however, increases in some regions appear more significant than others. Both NO_y-N and NH_x-N deposition rates increase most in the middle of mainland and southeast China, especially in the Yangtze River basin. There is a significant N deposition increasing gradient from the north and the northwest to the southeast, which is accordant with the N deposition distribution pattern in China (Fig. 3a and b). The average NO_y-N deposition rate increased from 1.25 kg N·ha^{-1}·yr^{-1} in the first decade to 3.40 kg N·ha^{-1}·yr^{-1} in the last decade, totaling an increase of 2.06 Tg N/yr; while the NH_x-N deposition rate is greatly increased on average, increasing from 2.72 kg N·ha^{-1}·yr^{-1} in the first decade to 15.44 kg N·ha^{-1}·yr^{-1} in the last decade, with a total difference of 12.17 Tg N/yr over the whole study period. In other words, atmospheric N deposited on the land surface of China has increased fivefold from 1901 to 2005 and the increase is dominated by increases in NH_x-N deposition. The annual increase rate of N deposition since 1950 is averaged to 0.19 kg N·ha^{-1}·yr^{-1}, more rapid than that in the first half of the 20th century, 0.08 kg N·ha^{-1}·yr^{-1} (Fig. 3c).

Considering the provincial boundary and contrasting climatic conditions, we divided China into five regions (Fig. 3): northwest (NW), mid-north (MN), northeast (NE), southwest (SW), and southeast (SE). Water deficit and low temperature are the major stresses for the NW, MN, and SW regions, while intensive human activities, such as land use and land management, are among the most dominant factors in NE and SE regions. N deposition level and its increase rate varied from region to region. N deposition increase peaked in southeast China, summing to 35.6 kg N/ha over the past 105 years, followed by north, southwest, northeast, and northwest, increasing by 17.4, 15.1, 14.7, and 7.6 kg N/ha, respectively. In addition, the major biomes in China have been exposed to different levels of N deposition and experiencing distinguished increases in N inputs. The N deposition increases in forest, cropland, and shrubland are very similar, totaling up to 26.6, 25.6, and 25.1 kg N/ha, respectively, over the study period, while increased N deposition is less significant in grassland with a value of 10.6 kg N/ha.

Other input data sets.—Besides N deposition data, three major groups of data sets were developed to characterize the environmental changes in China's terrestrial ecosystems over the past 105 years: (1) time-series data without spatial heterogeneity, e.g., atmospheric CO_2 concentration; (2) spatially explicit data without temporal variation, such as topography (DEM, slope, aspect), soil properties (depth, texture, bulk density, pH) and plant biogeographical features; (3) data with both spatial and temporal variations, including climate (maximum, minimum, and average temperature, precipitation, radiation, humidity), O_3 AOT40 index (an index of the accumulated hourly ozone concentrations above a threshold of 40 ppb·h), land use and land management practices (harvest, rotation, irrigation, fertilization). In this study, the resolution of all the spatial data is 10 km × 10 km, while the time steps for the above inputs range from daily (e.g., climate, O_3 AOT40 index) to yearly (e.g., CO_2 concentration, land use and land management patterns). DLEM model is run at daily time step. Except land use change and CO_2 concentration, all the annual input data sets have been interpolated into a daily step (e.g., N deposition, N fertilization; details can be found in the Appendix). Daily climate data during 1961–2005 was obtained by extrapolating from a monitoring network covering 740 weather stations in mainland China, six stations in Taiwan, and 29 stations from neighboring countries (Chen et al. 2006, Tian et al. 2011*b*, *c*). Climatic conditions from 1901 to 1960 were represented by the detrended climate data during the period 1961–1990, while climate in 1900 was depicted by using the mean daily climate data from 1961 to 1990. The atmospheric CO_2 concentration data from 1900 to 2005 was from the Carbon Dioxide Information Analysis Center (CDIAC) at Oak Ridge National Laboratory (Oak Ridge, Tennessee, USA). The soil depth, pH, texture maps, and chemical properties (Shi et al. 2004, Tian et al. 2010*b*) were generated based on the 1:1 million

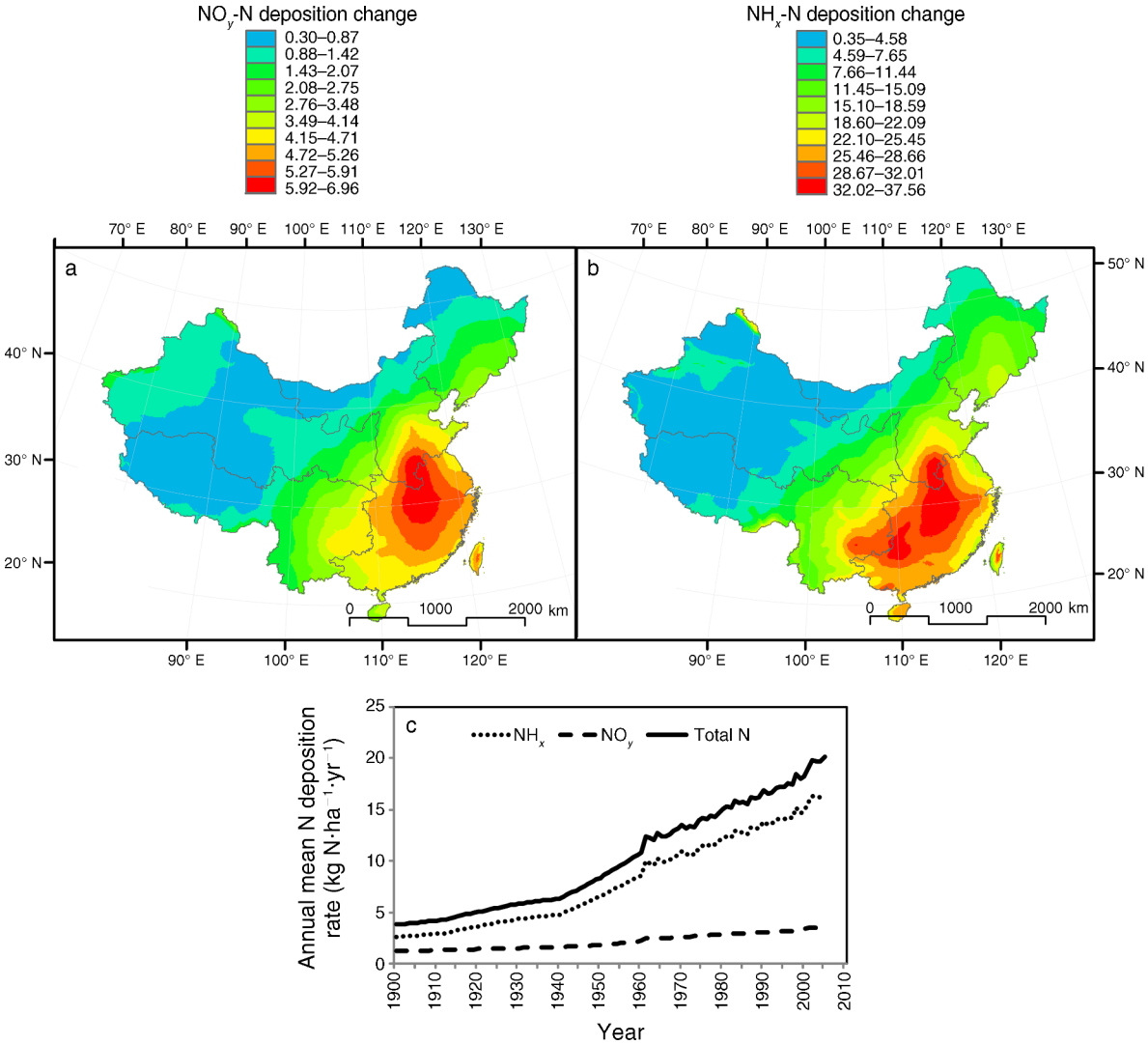

FIG. 3. Spatial pattern of N deposition changes (N dep. change; kg $N \cdot ha^{-1} \cdot yr^{-1}$) from the first decade (1901–1910) to the last decade (1996–2005) for (a) NO_y-N deposition and (b) NH_x-N deposition. (c) Time series of annual average N deposition rate in China during 1900–2005.

soil type map (Institute of Soil Science, Chinese Academy of Sciences, Nanjing, China) and the second national soil survey of China (National Soil Survey Office, Beijing, China). The O_3 AOT40 index from 1900 to 2005 was derived from a global historical AOT40 data set (Felzer et al. 2005) and validated against field observation in China (Ren et al. 2007). Spatially explicit land use history during 1900–2005 was retrieved from high-resolution remotely sensed data (Landsat TM/ETM+ and SPOT VEGETATION), field survey and the contemporary land use and land cover pattern indicated by China's National Land Cover Dataset (NLCD; Liu et al. 2005*a*, *b*, Liu and Tian 2010). Fig. 4 shows the contemporary vegetation map in the year of 2005 considering cropland establishment/abandonment and urban sprawl. We also developed an irrigation map, historical N fertilization map and cropping system information for managed cropland (Ren et al. 2011*b*, Tian et al. 2011*b*).

The input data sets show that significant changes in climate, atmospheric composition, land use, and management practices have taken place in China over time and space (Fig. 5; Tian et al. 2011*b*, *c*). During 1961–2005, the annual precipitation amount increased by 9.68 ± 4.06 mm per decade (mean ± SE) and the highest increase occurred in southeast China, while average air temperature rose at a rate of 0.29° ± 0.03°C per decade with northeast China experiencing the highest temperature increase (Tian et al. 2011*b*). Atmospheric CO_2 concentration was increased from 296 ppm in 1900 to 380 ppm in 2005, but our assumption of its homogeneous distribution over space would to some extent bias

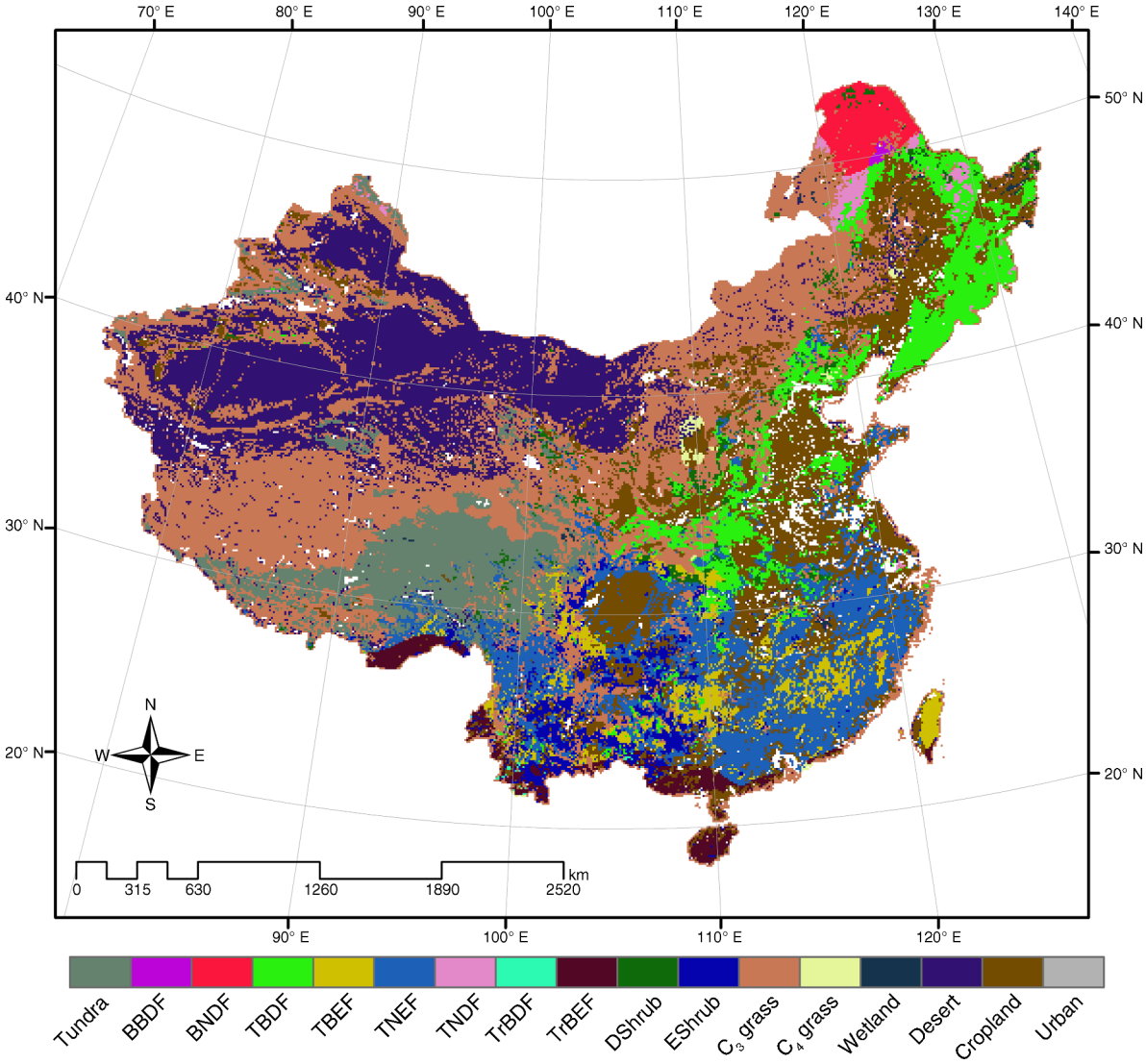

FIG. 4. Contemporary vegetation map of China in the year of 2005. Abbreviations are: BBDF, boreal broad-leaved deciduous forest; BNDF, boreal needle-leaved deciduous forest; TBDF. temperate broad-leaved deciduous forest; TBEF, temperate broad-leaved evergreen forest; TNEF, temperate needle-leaved evergreen forest; TNDF, temperate needle-leaved deciduous forest; TrBDF, tropical broad-leaved deciduous forest; TrBEF, tropical broad-leaved evergreen forest; Dshrub, deciduous shrubland; EShrub, evergreen shrubland.

the local estimates of N deposition effects. O_3 AOT40 index ranged from 0 to 3499 ppb·h over the study period and the increase rate became faster since the early 1990s due to rapid urban sprawl. Cropland in China expanded before the 1950s and then slightly decreased (Fig. 5e). Net increase of cropland area totals to 21.7 million hectares over the past 105 years and over half of the cropland establishment took place in northeast China (Liu and Tian 2010). On the national scale, forest area increased since 1949. Plantations largely contributed to forest expansion during the recent decades, however, a decline in forest area was found in northeast China (Liu and Tian 2010). N fertilizer application rate in agricultural land increased from 0 in 1900 to 22.23 g $N \cdot m^{-2} \cdot yr^{-1}$ in 2005. The highest fertilization rate was found in southeast China.

Simulation experiment design

Based on spatially explicit N deposition data and process-based ecosystem model with fully coupled C–N module, we set up three simulation experiments that aimed at taking the interactive effects between N input and other environmental changes into account (Table 4): (I) all-combined experiment, in which all driving factors are subject to change during 1901–2005; (II) combined without N deposition change, in which all

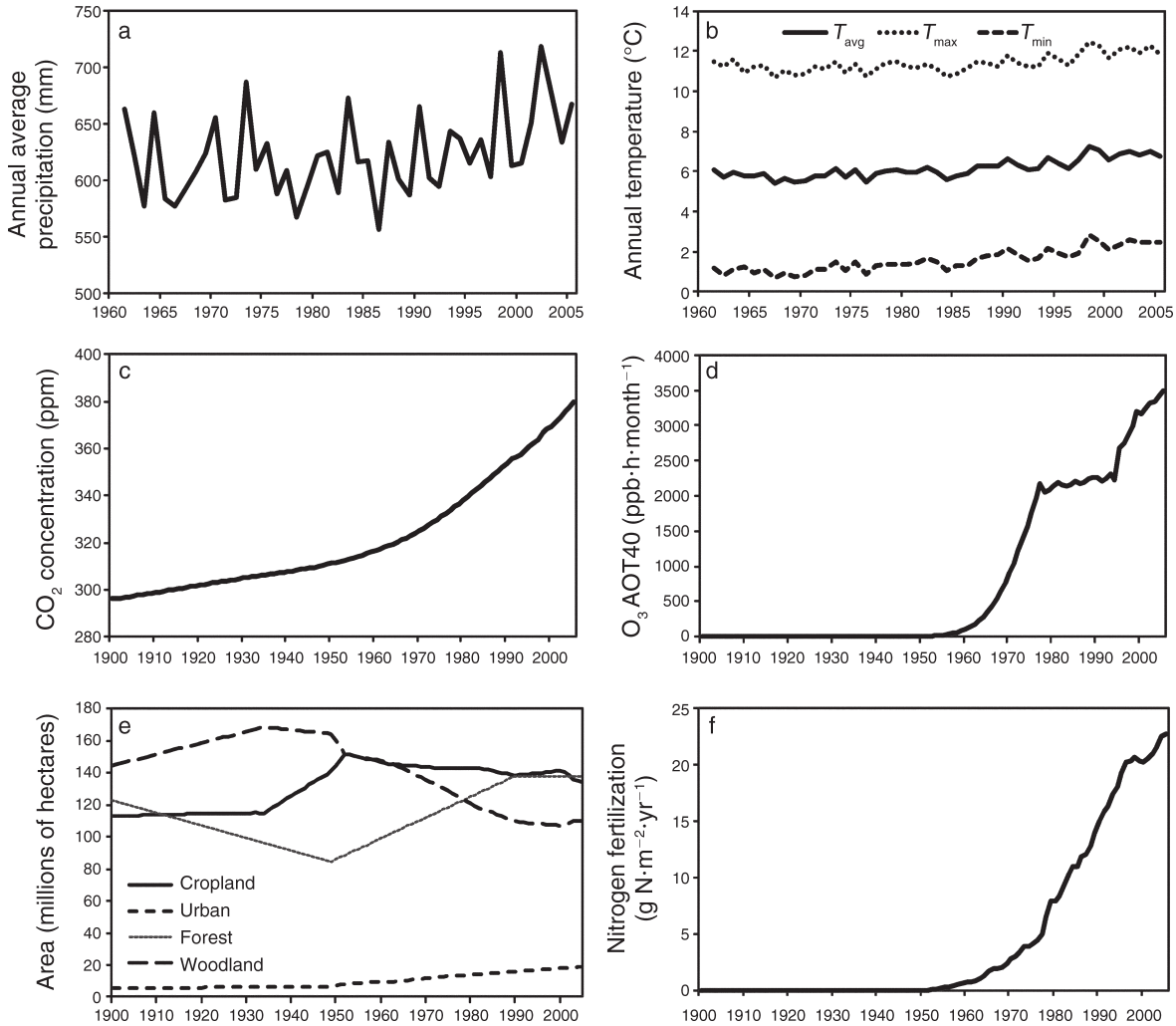

FIG. 5. Temporal variations in (a) annual mean precipitation (mm) over the period 1961–2005; (b) annual mean (T_{avg}), maximum (T_{max}), and minimum (T_{min}) temperature (°C) during 1961–2005 (before 1961, climate conditions were represented by the detrended climate data during the period 1961–1990); (c) atmospheric CO_2 concentration (ppm); (d) annual mean monthly O_3 AOT40 index (ppb·h·mo^{-1}; an index of the accumulated hourly ozone concentrations above a threshold of 40 ppb·h); (e) area of land cover types (millions of hectares); (f) annual average N fertilizer application rate (g N·m^{-2}·yr^{-1}) in China during 1900–2005.

factors change except N deposition remains at the level of the year 1900; (III) N deposition only, in which all factors are held constant at the level of the year 1900 except that N deposition changes from 1901 to 2005. In addition, in order to examine how CO_2 fertilization affected the simulated response of terrestrial C dynamics to atmospheric N addition in China, we conduct two more simulation experiments (experiments IV and V in

TABLE 4. Design of simulation experiments.

Number	Experiment	Climate	CO_2	O_3	Ndep	Nfer	LUCC
I	All combined	1900–2005	1900–2005	1900–2005	1900–2005	1900–2005	1900–2005
II	Combined without Ndep change	1900–2005	1900–2005	1900–2005	1900	1900–2005	1900–2005
III	Ndep only	1900	1900	1900	1900–2005	1900	1900
IV	Combined without CO_2 change	1900–2005	1900	1900–2005	1900–2005	1900–2005	1900–2005
V	Combined without Ndep and CO_2 change	1900–2005	1900	1900–2005	1900	1900–2005	1900–2005

Notes: Ndep, Nfer, and LUCC are abbreviation for N deposition, N fertilization and Land use and cover change, respectively. For the experiments I, II and III, two different N deposition data sets have been used to drive the DLEM model: one is developed in this study; the other is from Dentener et al. (2006). Date ranges indicate that the environmental factors (column heads) in those time periods were used to drive the model.

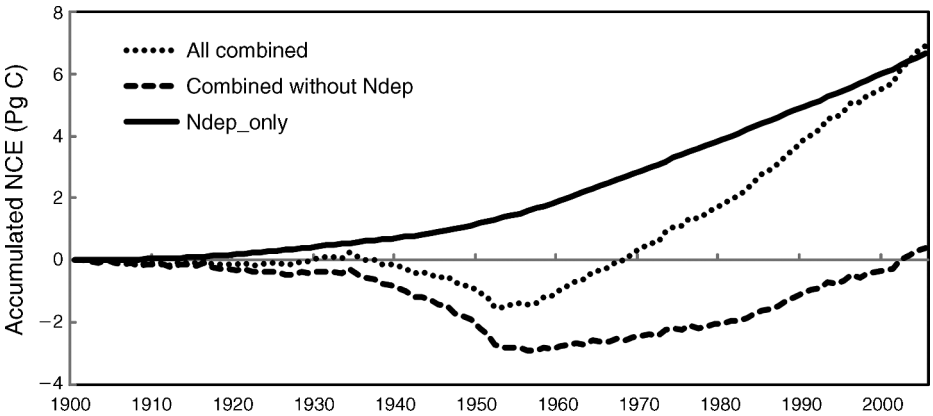

FIG. 6. Accumulated net carbon exchange (NCE) induced by changes of nitrogen deposition alone (Ndep_only, solid line), combined changes of all factors (all combined, dotted line), and all factors' changes except nitrogen deposition (combined without Ndep, dashed line) from 1900 through 2005. The difference between the latter two is the impact of nitrogen deposition change and its interaction with other factors.

Table 4) in which atmospheric CO_2 concentration was kept constant at the level of 1900 while other environmental factors remained the same as the experiments of "all combined" and "combined without N deposition change," respectively.

We assume 1900 represents a preindustrial era and that anthropogenic N deposition at that time is low enough to be ignored. Experiment II is identified as a control experiment representing the C cycle unaffected by N deposition change. Therefore, the difference between experiments I and II reflects the effects of increased atmospheric N input and its interactive effect with other environmental changes, while the third experiment indicates the response of C fluxes and storage to N deposition change alone. In this study, the response values, which we estimate to evaluate the extensive impacts of increased N deposition, refer to the differences of C variables (ΔC) between experiments I and II. Furthermore, the ratio (%) of ΔC to C variable value in the control experiment is calculated as a response ratio. Through this standardized term, we can compare the magnitude of responses among different variables and biomes. The response ratio is an indicator of N deficit in determining ecosystem C dynamics. The larger the ratio is, the more N limitation to a certain variable can be alleviated by atmospheric N inputs.

Before each transient run, the model was run to equilibrium, defined as the year-to-year changes in NCE, soil water content, soil mineral N content, and plant N uptake are less than 0.1 g C/m^2, 0.1mm H_2O, and 0.1 g N/m^2, respectively, over 10 consecutive years. Using atmospheric CO_2 concentration, O_3 AOT40 index, land use types, and land management practices in the year of 1900, DLEM was run to reach equilibrium, which is used to determine the initial conditions of the simulated C dynamics. After that, a spin-up of 100 iterations using the values of atmospheric composition, land use, and land management in 1900 is performed to return the model to dynamic equilibrium. For the spin-up, detrended climate data from 1901 to 1930 were used for experiments including transient climate change, while mean daily climate data from 1901 to 1930 was used for other experiments. After a 100-year spin-up, the model was fed by the time series of input data set in transient mode.

TABLE 5. Responses of (a) C fluxes (Pg C/yr) and (b) C pools (Pg C) to N deposition change in China's terrestrial ecosystems during the past 105 years

a) C fluxes:

	ΔNPP		ΔR_h	
Decade	Mean	%	Mean	%
1900s	0.01	0.28	0.00	0.09
1940s	0.12	4.88	0.06	2.71
1960s	0.29	11.43	0.16	6.88
1980s	0.40	12.98	0.25	9.86
2001–2005	0.45	13.01	0.31	11.60

b) C pools:

	ΔVegetation C		ΔLitter C		ΔSoil C		ΔTotal C	
Year	Mean	%	Mean	%	Mean	%	Mean	%
1901	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1941	0.40	1.52	0.05	1.67	0.26	0.35	0.71	0.69
1961	0.96	3.88	0.13	4.31	0.73	1.01	1.82	1.81
1981	1.91	7.63	0.27	8.83	1.65	2.25	3.84	3.78
2005	3.10	11.83	0.46	14.50	2.87	3.87	6.43	6.22

Notes: The symbol Δ indicates changes in C fluxes and storage. The "Mean" columns report the average N-induced C dynamics in each time period. Percentage (%) is calculated as the ratio of N-induced change in carbon fluxes or carbon storage to the corresponding level in the control experiment, multiplied by 100. The decade 1900s refers to the period 1901–1910; 1940s, 1960s, and 1980s refer to the periods 1941–1950, 1961–1970, and 1981–1990, respectively.

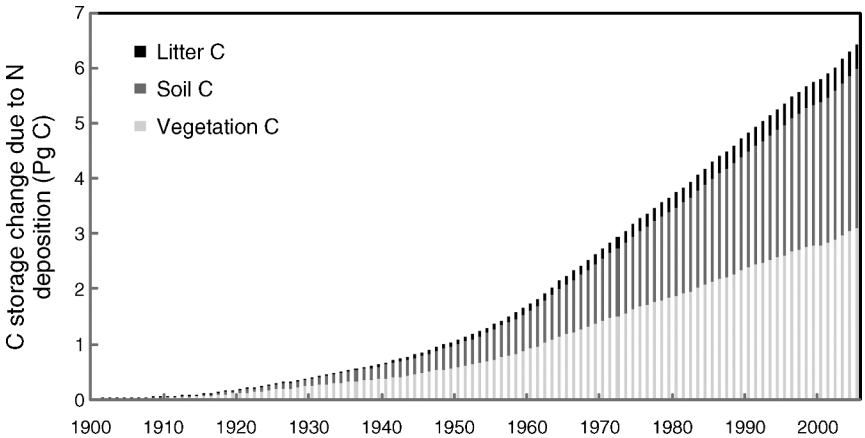

FIG. 7. Carbon storage change among vegetation, litter, and soil pools due to nitrogen deposition change and its interaction with other environmental factors over the past 105 years.

RESULTS

Response of C fluxes and storage to N deposition on national level

Both the simulation approaches with and without consideration of interactive effects reveal that the annual N-induced C sequestration capacity has been augmented with the elevated N deposition levels over time (Fig. 6). Relative to the control experiment without N deposition increase, N-induced changes in C fluxes and storage demonstrate an increasing gradient from 1901 through 2005 (Table 5). Our model simulation indicates that, across China, both net primary productivity (NPP) and heterotrophic respiration (R_h) increased in response to atmospheric N inputs over time, but the magnitude and percentage change of altered NPP were larger than those of altered R_h during the same period. Hence, N addition has resulted in a net C sink of 62 Tg C/yr as simulated by DLEM, and accumulated 6.51 Pg C over China's terrestrial ecosystems during 1901–2005. Compared to the CO_2 emission rate reported by Boden et al. (2009), the modeled terrestrial C sequestration due to elevated N deposition can compensate for more than 25% of fossil-fuel CO_2 emission from China over the past 105 years. Although plant growth is largely promoted by enhanced N availability in extensive areas of China, the consequent C uptake still falls far behind the accelerated C emissions from various anthropogenic sources. For example, over the period 1996–2005, the modeled annual average N-induced C uptake only made up for 10% of the C release, which has been increased substantially in recent decades.

Excluding the C allocated to product pool and microbe biomass, the N-induced C storage change is estimated to be 6.43 Pg C in China over the entire study period. Our estimation shows that N-induced C storage amounts to 1.74 Pg C from 1901 to 1960, which accounts for only 27% of the C accumulation during the past 105 years. However, C storage over the recent 45 years is estimated to be 4.69 Pg C, nearly three times the amount of the C stored in the prior 60 years. The DLEM model estimates that N deposition combined with its interactive effects contributed to 45% of net carbon storage driven by changes in climate, atmospheric composition (CO_2, tropospheric O_3, N deposition), land use/land cover patterns and agricultural management practices (e.g., harvest, rotation, irrigation, fertilization) in China during 1961–2005. By incorporating model estimates from an additional model, the terrestrial ecosystem model (TEM), our previous study has the same conclusion (Tian et al. 2011*b*).

We find that the increasing rate of C storage has been greatly increased since the 1950s, which is largely related to the rapid N deposition increase over this time period (Fig. 3c and Fig. 7). The vegetation C pool, particularly in forest ecosystems, shows significant response to increased N input due to its higher C:N ratio. Of the storage compartments, living vegetation, soil, and litterfall account for 48%, 45%, and 7% of the resulting increase in total C pool size, respectively, in the year of 2005 (Table 5). In comparison, 53%, 40%, and 7% of C storage arising from N deposition has been allocated to vegetation, soil, and litterfall pools, respectively, during the previous 60 years. This implies that more C is prone to be allocated into vegetation with an early increase in N input, while continuous N addition can increase the proportion of soil C (Fig. 7). The increasing importance of soil carbon pool can be largely attributed to a modeled phenomenon that most of N deposited from the atmosphere is retained by soil and the pool size of soil N increases more rapidly than that of vegetation N as N input rises (Lu 2009), which is also shown by isotopic tracer and long-term N fertilization experiments (Nadelhoffer et al. 1999, Magill et al. 2000). Although most N is shown to be retained in the soil pool, the higher C:N ratio in plants than soils (C:N $>$ 300 in woody tissue, 10–30 in forest soil; Schindler and Bayley 1993, Towsend et al. 1996) leads to a large proportion of C storage allocated into vegetation.

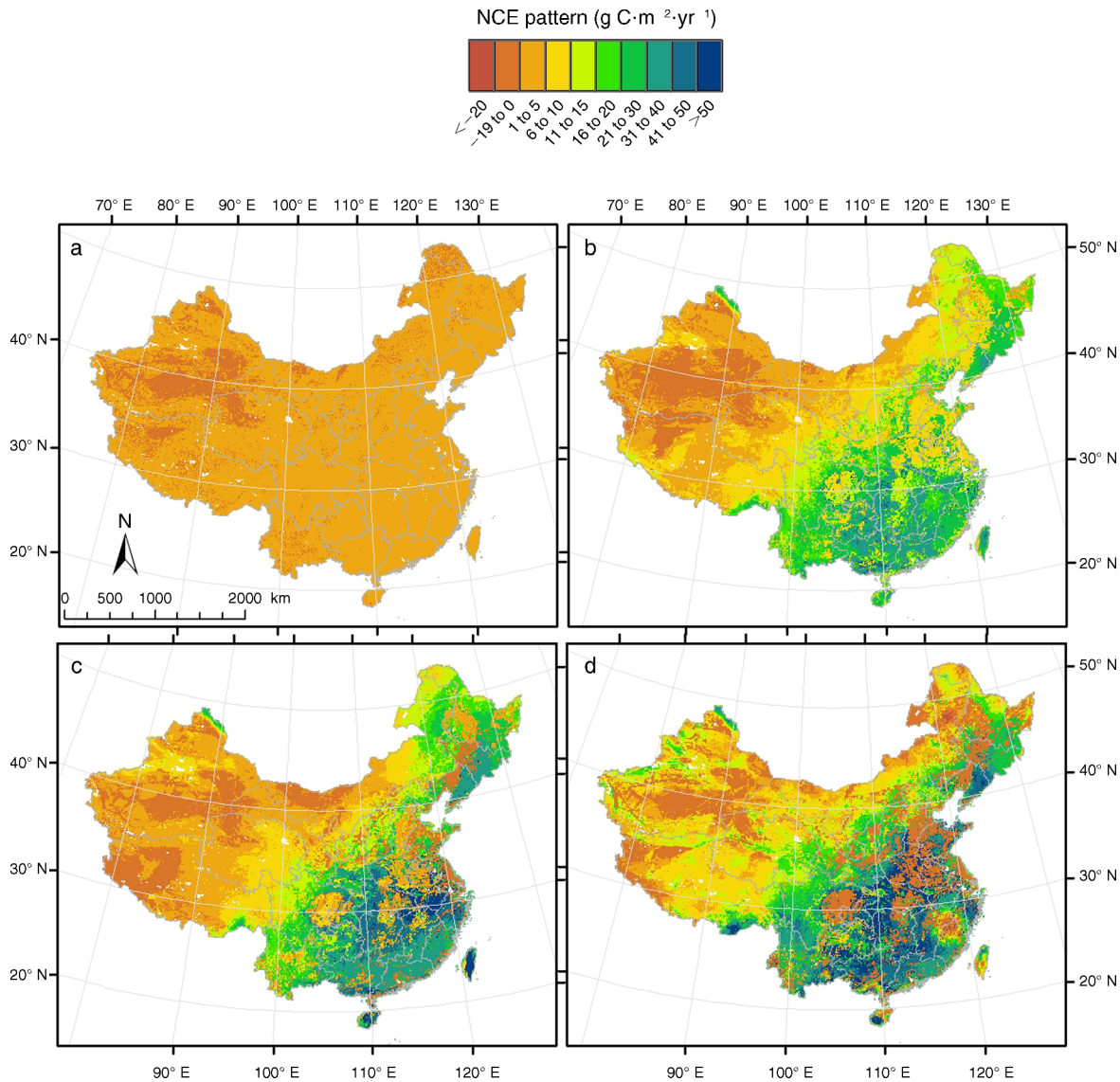

FIG. 8. Spatial distribution of net carbon exchange (NCE) resulting from increased N deposition in China during (a) the 1900s, (b) the 1960s, (c) the 1980s, and (d) 2001–2005. (NCE is calculated as the difference between experiments I all combined and experiments II combined without N deposition change in a certain decade.)

Nationwide, the C sequestration induced by N deposition presents a significant gradient from northwest to southeast China (Fig. 8), which agrees well with the spatial pattern of N deposition. Furthermore, with enhanced N deposition, the C sequestration capacity in southeast China has been greatly increased, even reaching more than 50 g $C \cdot m^{-2} \cdot yr^{-1}$ across some regions in the early 2000s (Fig. 8d), and the area of N-induced C sink in north and northeast China has been considerably enlarged from 1901 to 2005 (Fig. 8a–d). It is noticeable that the southeastern C sink was diminished, and even changed into a C source in the early 2000s, although the C uptake areas were still expanding toward the northern and northwestern regions. By analyzing the N-deposition-only experiment, however, we found that atmospheric N inputs continued to stimulate C sequestration in southeast China from the 1980s through the early 2000s (Fig. 9). Most grids characterized by an N-induced C source in the recent years coincide with the area covered by crops. Land management practices, such as harvest, rotation, irrigation, and N fertilizer application in agricultural land, would make the pattern of N effects even more complex. In the model assumption, not only photosynthesis but also residue and crop yield from land use and cover changes are stimulated by N additions. Our simulation shows that NPP enhanced by N enrichment was not large enough to offset the increased C loss due to respiration, land conversion, and consumption of crop products in these regions. Therefore, the conversion from C sink to source suggested by Fig. 8 can be mostly

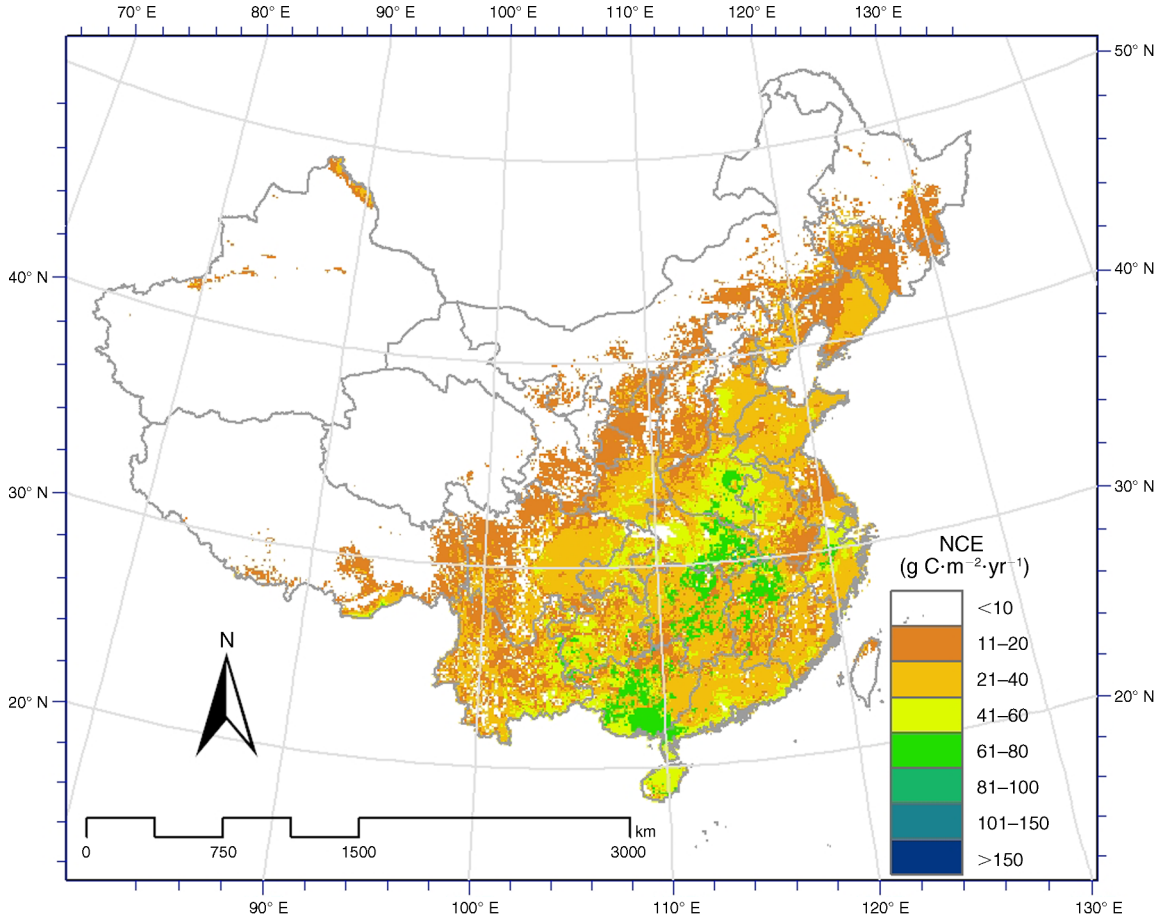

FIG. 9. Spatial pattern of altered net carbon exchange (NCE; g $C \cdot m^{-2} \cdot yr^{-1}$) due to nitrogen deposition change alone between the 1980s and the early 2000s.

attributed to the N effects regulated by other environmental changes, such as deforestation, cropland establishment, and management.

Biome response of C fluxes and storage to N deposition in China

Both C sequestration capacity and C allocation into storage pools vary significantly among the major biome types in China (Table 6). We grouped the key vegetation types into four categories: forest, shrubland, grassland, and cropland. From 1901 through 2005, model simulation indicates that forest and grassland were among the greatest contributors of N-induced C sequestration, with annual average NCE of 37 and 11 Tg C/yr, while shrubland and cropland only sequestered 4 Tg C/yr and 2 Tg C/yr, driven by increased N deposition. To summarize, forest accounted for 60% of N-induced C storage, followed by grassland, shrubland, and cropland, with contributions of 18%, 7%, and 4%, respectively. The rest 1% came from desert, tundra, wetlands, and others. Though the soil C pool plays an increasingly important role with enhanced N deposition level, as indicated above

TABLE 6. N deposition-induced carbon sequestration and the consequent carbon storage in major biomes of China during 1901–2005.

Biome	N-induced NCE ± SD† (Pg C/yr)	Accumulated NCE (Pg C)	Biome proportion	Vegetation C (Pg C)‡	Soil C (Pg C)‡	Litter C (Pg C)‡
Forest	0.037 ± 0.026	3.89	60%	2.06 (55%)	1.3 (35%)	0.4 (11%)
Grassland	0.011 ± 0.010	1.14	18%	0.47 (43%)	0.59 (54%)	0.03 (3%)
Shrubland	0.004 ± 0.003	0.48	7%	0.29 (62%)	0.18 (38%)	0.004 (1%)
Cropland	0.002 ± 0.005	0.25	4%	0.001 (0.4%)§	0.34 (99.4%)	0.001 (0.2%)

† SD is the standard deviation of inter-annual NCE change due to increased N deposition.
‡ The percentage is the proportion of each carbon pool relative to total C storage for a specific biome type.
§ Vegetation C induced by N deposited onto cropland is nearly 0 because most crop biomass has been removed by harvest and product C change is not accounted in this study.

TABLE 7. The simulated biome response of C dynamics to N deposition change in China's terrestrial ecosystem over 1901–2005.

Biome	NPP		R_h		Veg. C		Soil C		Total C	
	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)	Mean (%)	SD (%)
Boreal forest	3.92[a]	3.90	2.13[a]	1.82	1.70[a]	1.49	0.61[a]	0.65	1.17[a]	1.09
Temperate forest	7.82[b]	6.38	4.99[c]	4.57	3.61[c]	3.45	1.70[c]	1.86	2.75[c]	2.76
Tropical forest	4.26[a]	3.64	2.68[b]	2.48	2.19[b]	2.06	1.15[b]	1.24	1.76[b]	1.74
Broadleaf forest	7.70[b]	5.44	4.24[a]	3.86	3.01[a]	2.89	1.55[a]	1.69	2.40[a]	2.41
Needleleaf forest	6.73[a]	6.29	4.91[b]	4.52	3.67[b]	3.45	1.57[b]	1.72	2.66[b]	2.64
Forest	7.22[b]	5.86	4.58[a]	4.19	3.33[a]	3.16	1.56[b]	1.71	2.53[b]	2.53
Shrubland	13.19[d]	11.03	8.84[c]	8.22	11.88[d]	11.02	3.18[d]	3.42	6.13[d]	6.01
Grassland	6.60[a]	5.34	4.57[a]	3.97	6.06[c]	5.39	0.61[a]	0.66	1.14[a]	1.11
Crop	7.90[c]	5.88	5.80[b]	4.25	4.94[b]	4.03	2.86[c]	2.35	2.93[c]	2.35

Notes: Response (%) is calculated as the percentage change of carbon fluxes and carbon storage due to N deposition input relative to their controlled levels without N enhancement. SD is the standard deviation of inter-annual biome response to N deposition during 1901–2005. Different letters beside the values indicate significant differences of each variable within groups of boreal, temperate, and tropical forest groups of broadleaf and needleleaf forest and among forest, shrublend, grassland, and crops. "Veg." stands for vegetation.

(Fig. 7), C allocation patterns are distinct from biome to biome. For forest and shrubland, 55–62% of C storage increase induced by N inputs is estimated to allocate into the vegetation pool, while vegetation carbon only accounts for 43% of the total N-stimulated C stock in grassland. The C:N ratios of non-woody tissue in grassland are far less than that of woody tissue in the former two biome types; in addition, the higher turnover rate in grassland transfers more C into the soil pool. As for cropland, N-induced vegetation C pool can be ignored, large portion of which has been removed through harvesting, and therefore, most of C is stored in soil.

Over the past 105 years, responses of C dynamics to N deposition show large divergence among biomes (Table 7). We find that shrubland shows the largest response ratio to N deposition increase, followed by cropland, forest, and grassland. However, considering the absolute change values, the NPP in shrubland was increased by 0.01 Pg C/yr due to atmospheric N deposition, which is nearly one-third of N-induced NPP in grassland and cropland, and one-tenth of NPP increase in forest as simulated by DLEM. This implies that the N deficit in shrubland was more relieved by atmospheric N inputs than that in other biomes of China, even though shrubland has the least N-stimulated productivity. The magnitude of this relief is determined by the combination of initial N content, ecosystem demands and amount of N input. It's hard for us to identify which factor dominantly contributes to the higher response in shrubland relative to other biomes.

In order to examine the biome-level NCE in response to N deposition change alone, we examined the spatial variation of C sequestration change between the 1900s and the early 2000s by using the N-deposition-only experiment (Fig. 10), which excluded impacts from other environmental stresses. Over the past 105 years, the C sink in forest ecosystems has been greatly enlarged by N deposition increase nationwide. N-induced NCE peaked in the central part of mainland and southeast China, as well as small area of the northeast, even reaching 50 g $C \cdot m^{-2} \cdot yr^{-1}$. By contrast, large areas of grassland sequestered less than 10 g $C \cdot m^{-2} \cdot yr^{-1}$ in response to deposited N enrichment. This difference is partly because most grassland ecosystems are distributed in high-latitude areas or arid to semiarid regions, where plant growth is more limited by low temperature and/or low soil moisture than by N availability. In addition, the low increase in the rate of N deposition also contributes to the smaller response of grassland. NCE change in shrubland presented two distinguished patterns. Due to spatial heterogeneity of N deposition, shrubland in southwest China took up C at a rate of more than 10 g $C \cdot m^{-2} \cdot yr^{-1}$, while that distributed in northwest and mid-north region sequestered C at a much lower rate. Given moderate or higher N deposition level to which cropland was exposed, atmospheric N enrichment caused a less C sink in agriculture ecosystems throughout the study period. As we know, deposited N from the atmosphere may not further stimulate crop growth like it does in other ecosystems because N fertilizer application and cultivated legumes have already provided adequate exogenous N (Townsend et al. 1996). In China particularly, a recent study (Lu et al. 2009) has documented that N fertilizer application can sequester 5.96 Tg C in soils of rice, wheat, and maize in China each year and it has been further confirmed that fertilized N has largely surpassed the plant demands in cropland and consequently triggered a series of environmental problems (Ju et al. 2009).

N saturation issue in China

In this paper, we use N-induced NPP relative to N deposition inputs to indicate the ecosystem N use efficiency (ENUE). In an N-limited ecosystem, ENUE tends to increase with N enrichment as N deficit has been to some extent alleviated, for example, ENUE in China increased during the early past century (Fig. 11). However, the model simulation shows that NPP and R_h changes in response to per-unit N deposition began to level off or even decline since the 1980s, although the N

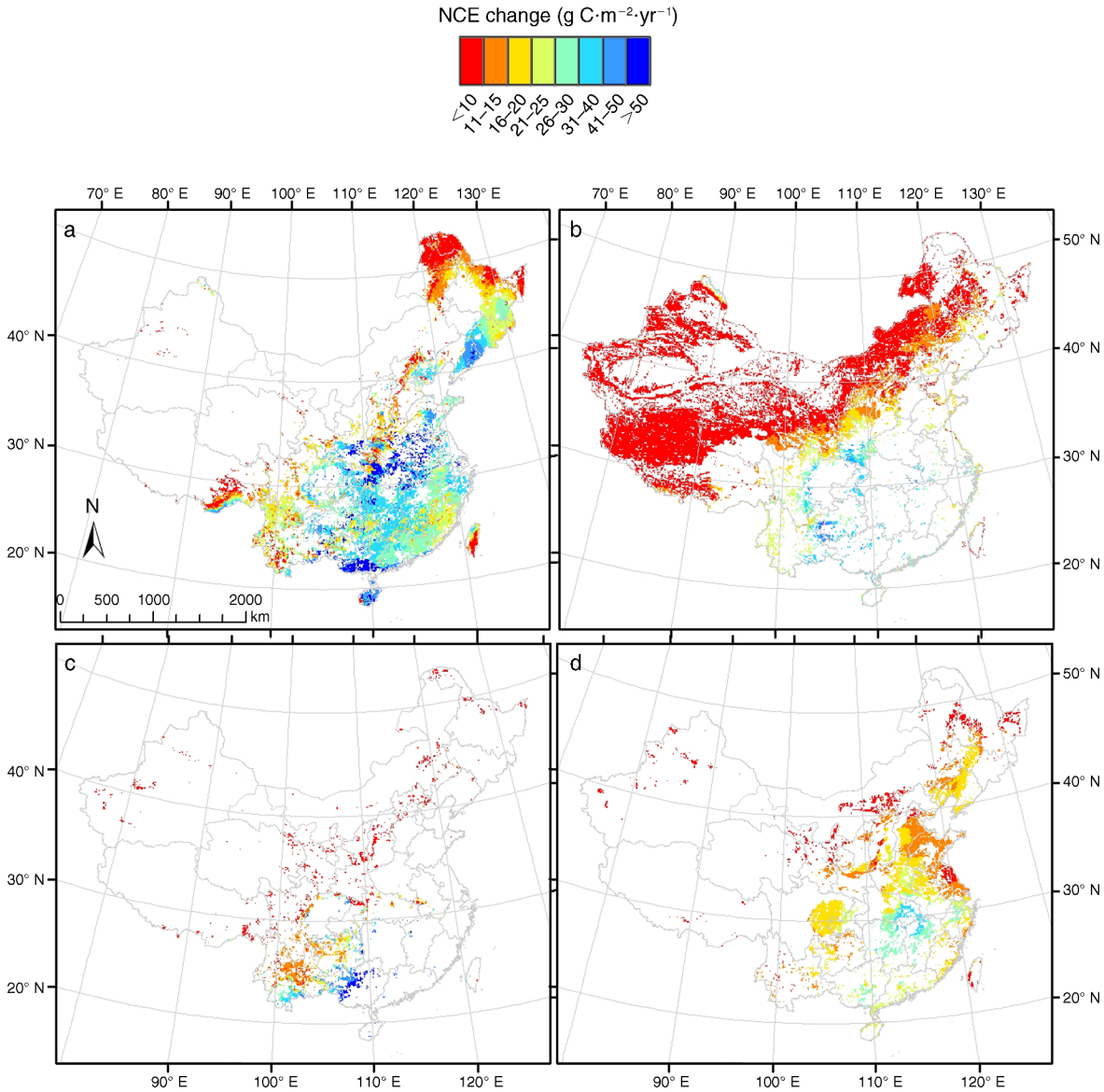

FIG. 10. Spatial pattern of net carbon exchange (NCE) change in response to N deposition change alone over major biomes of China during the 1900s to early 2000s: (a) forest, (b) grassland, (c) shrubland, (d) cropland.

deposition level continued to increase (Fig. 11). Plant N use efficiency (PNUE, i.e., the ratio of NPP to plant N uptake) is found to be relatively constant for a certain biome type (Lu 2009). Therefore, reduction in ENUE indicates that part of exterior N inputs is not retained in the system to further stimulate plant growth, but leaves the ecosystem through DIN and DON leaching, nitrous gas emissions, and other pathways. This phenomenon reflects that N retention capability in China's terrestrial ecosystems has reached its summit and that the additional N cannot be fully or efficiently invested to sequester C, which we recognize as an evidence of nitrogen saturation; that is, a condition in which nitrogen inputs to an ecosystem exceed the demands of plants and microbes. As Aber et al. (1998) stated, one of critical thresholds during N saturation process occurs when nitrogen becomes a non-limiting element in plant growth and, above this point, detrimental consequences including increased NO_3^- leaching loss and N_2O efflux will be caused by excessive N input. A site-level experiment conducted in southeastern China (Fang et al. 2008) showed that N addition didn't stimulate plant growth, especially for mature forest, and that fertilized N was not retained in the soil, on the contrary, there was a net loss of 8–16 kg $N \cdot ha^{-1} \cdot yr^{-1}$ with wet N deposition of more than 50 kg $N \cdot ha^{-1} \cdot yr^{-1}$.

During 1901–2005, ENUE showed different patterns among five regions in China. Except northwest and

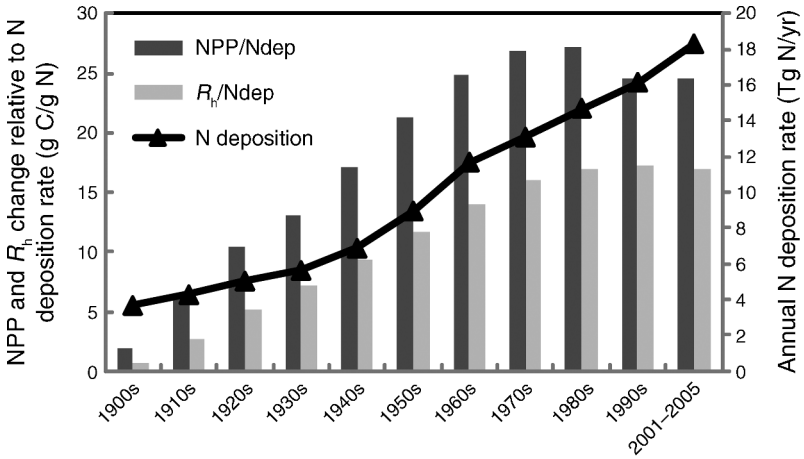

FIG. 11. Decadal average net primary production (NPP) and soil respiration changes (heterotrophic respiration, R_h) induced by per-unit nitrogen deposition in China from 1901 to 2005.

southwest China, the stimulation of deposited N to NPP reached its peak in the other three regions around the 1980s (Fig. 12a), where atmospheric-deposited N was substantially enhanced. In terms of the two non-saturated regions, the northwest region had the lowest N input level and the slowest N addition rate; whereas the southwest region experienced moderate N deposition, but its ENUE continued to increase. Southwest China ranked as the second greatest N-induced C pool, accounting for 25% of the total C stock due to atmospheric N enrichment, while the N deposition level of southwest China was nearly half of the level in southeast China and the latter contributed to 39% of the N deposition-induced C storage. The model simulation indicates that the ecosystem in the southwest is not N saturated yet.

Likewise, ENUE showed distinguished patterns among biomes. Since the 1980s, N-induced NPP relative to per-unit N deposition began to level off in all biomes except shrubland (Fig. 12b). As we discussed before, the N deficit in shrubland has been largely alleviated by enhanced N deposition but not enough to shift the ecosystem into N-saturated status yet. It is noticeable that ENUE in cropland has been reduced since the 1960s, and finally reached a level close to that in the 1900s. Since agriculture management practices such as fertilizer application and irrigation have been widely promoted since the 1960s, only a small portion of deposited N has been retained and used for plant growth. In recent and coming decades, atmospheric N-induced NPP in cropland was and will continue to be the lowest of all the biomes.

DISCUSSION

Impacts of N deposition on C fluxes and storage

The existing simulation studies on N deposition effects show substantial variability in their estimations of net carbon exchange between land ecosystem and the atmosphere. During the 1990s, these model results of C sink range from 0.44–0.74 Pg C/yr driven by N input from global fossil fuel combustion (Townsend et al. 1996), to 0.2–2.0 Pg C/yr due to anthropogenically increased N deposition on the globe (Gifford et al. 1996, Holland et al. 1997, Jain et al. 2009) and 0.25–2.21 Pg C/yr in temperate forests (Nadelhoffer et al. 1999, Churkina et al. 2007). Only a few of them include spatially explicit information about N deposition in examining the distribution of C sinks (Townsend et al. 1996, Holland et al. 1997, Churkina et al. 2007, Jain et al. 2009). Here, C sequestration due to N deposited onto China's terrestrial ecosystems is estimated by DLEM to be 0.094 Pg C/yr over the same period, accounting for 5–47% of the above-reported global C uptake.

Various studies showed large divergence in estimating the size of C sink induced by per-unit N input. For example, Townsend et al. (1996) estimated an additional land C sink of 0.44–0.74 Pg C/yr due to 17 Tg fossil-fuel N deposited in the year of 1990, implying a net sequestration of 26–44 kg C/kg N deposited. Holland et al. (1997) showed an even higher estimate of 1.5–2.0 Pg C/yr induced by N deposition resulting from human activities, including fossil fuel combustion, biomass burning, and N fertilizer application in the form of NO_y-N (15.4–27.6 Tg N/yr) and NH_x-N (22.5 Tg N/yr) in the same year, with C response to N deposition ranging from 30 to 71 kg C/kg N. Nadelhoffer et al. (1999) conducted a series of ^{15}N-isotopic tracer experiments and found that 70% of the additional N was assimilated by soil with lower C:N ratios, therefore, the annual atmospheric N input of 5.1 Tg N made a minor contribution to temperate forest C sink in the order of only 0.25 Pg C/yr, suggesting a response ratio near 50 kg C/kg N. However, Jain et al. (2009) suggested that the atmospheric input of 62.2 Tg N/yr led to a land C sink of 0.26 Pg C/yr with a lower response rate of 4 kg C/kg N during the 1990s. A recent study indicates that the response ratio ranges from 5 to 75 kg C/kg N for

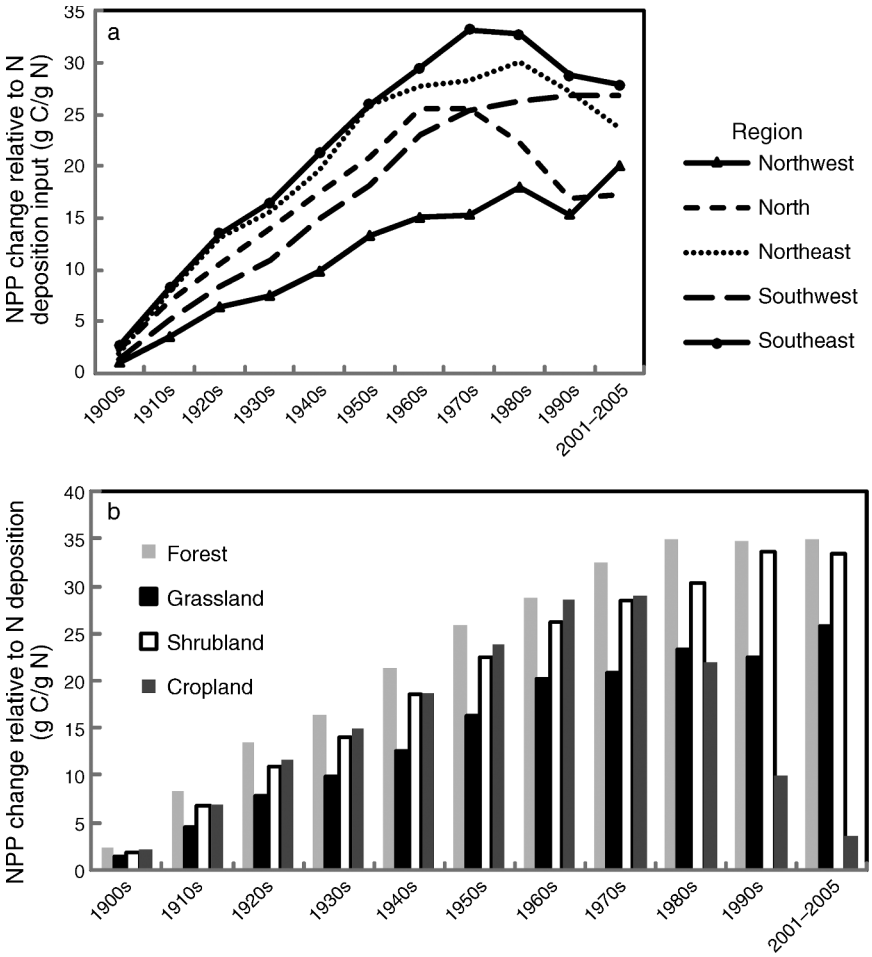

FIG. 12. Decadal change in net primary productivity (NPP) in response to per-unit N deposition input among (a) five regions and (b) major biomes in China during 1901–2005.

European forest and heathlands and large uncertainties still exist in C sequestration induced by per-unit N deposition (De Vries et al. 2009). Our simulation indicates a response of around 0–21 kg C/kg N over whole nation in the 1990s, falling on the lower end of the above response ranges, which might be related to the inclusion of less responsive ecosystems, such as grassland, cropland as well as desert, and the fact that lower ENUE occurred in response to high N input levels.

The DLEM-simulated NPP change was shown to be consistent with the observed response in face of high-level N additions in multiple manipulated experiments (see *Methods and model strategy: Model calibration and validation*), however, chronic N deposition increase in the real world are not expected to affect NPP to the same extent as N fertilization experiments suggested. As Högberg et al. (2006) argued, high doses of N addition over short periods do not mimic the long-term effects of N addition at lower rates. Although a wide range of simulation studies paid much attention to assessing how N deposition change affected terrestrial C balance, the magnitude of C flux responses to atmospheric N enrichment are still under debate. Few experiments have been carried out to investigate the ecosystem responses to ambient N deposition. It hinders the verification of model behavior in depicting long-term N effects. For example, according to our model simulation, the increased NPP, in response to average N enrichment of 19.8 kg $N \cdot ha^{-1} \cdot yr^{-1}$ during the last five years, was equivalent to 13.01% of NPP in the control experiment (Table 5). By contrast, the meta-analysis involving 126 N-addition experiments in various ecosystem types argued that the ratio of N-induced aboveground net primary productivity (ANPP) change relative to ANPP in control plots averaged 29% with N inputs level of 143 $\pm$ 11 kg $N \cdot ha^{-1} \cdot yr^{-1}$ (mean $\pm$ SE; LeBauer and Treseder 2008). It is hard to tell whether the ecosystem model ought to be improved in its assumptions and equations, or whether this inconsistency is caused by the inherent variability in terrestrial ecosystems' responses to short-term high-dose and long-term low-dose N supply. Therefore, the field experiments involving long-term multiple levels of ambient N deposition are more than urgent for model development and evaluation.

Likewise, the simulated biomass response ratio to ambient N deposition ranges from 0% to 11.83% (Table 5), which is considerably smaller than that recently reported at the species level (Xia and Wan 2008). They pointed out that the natural-log-transformed ratios of plant biomass at elevated and ambient N levels are 50.7% and 24.6% for herbaceous and woody species, respectively, equivalent to 66% and 28% in the response ratios we defined. The large discrepancy between responses to N addition on the community level (Elser et al. 2007, LeBauer and Treseder 2008) and the species level (Xia and Wan 2008) calls for more future work focusing on N impacts from the ecosystem perspective. It's also critical for better understanding the ecosystem behaviors in response to chronic atmospheric N enrichment and improving the regional model construction.

We also find that more C is sequestrated into vegetation pool than in soil and litter pools due to atmospheric N enrichment (Table 5). The percentage of C storage change relative to controlled C stock implies that vegetation and litter C are more sensitive than the soil C pool to elevated N availability. This pattern might be partly associated with the higher C:N ratios in vegetation and litterfall. On the other hand, as the isotopic tracer experiment and long-term fertilization experiments revealed, more added N has been retained in soil and partly mitigated its N limitation to the formation of soil organic matter (Tietema et al. 1998, Nadelhoffer et al. 1999, 2004, Magill et al. 2000). Our model simulation also confirms that nearly 40% of N deposition was retained in the terrestrial ecosystems of China over the recent decades and more than 85% of the total N storage increase resulting from atmospheric N input was found in the soil pool. The above two reasons probably contribute to the less responsiveness of soil C stock to N additions than vegetation and litter pools.

The model simulation reveals that growth of temperate forest shows the greatest response ratio to atmospheric N input, followed by tropical forest and boreal forest (Table 7). Undoubtedly, temperate forest ecosystems have been experiencing the greatest increase in N deposition over the past decades, by which N stress has been alleviated to a great extent. Table 7 indicates that the differences of simulated NPP in response to N enrichment between tropical forest and boreal forest are insignificant, which is partly caused by co-limitation from other environmental factors. Matson et al. (1999) pointed out that increasing atmospheric N inputs are unlikely to increase productivity of tropical forests where N is abundant and other resources are in short supply. Likewise, boreal forests might be less responsive to N input when temperature and water are more limiting to plant growth relative to N availability. However, over a long period, regional simulation didn't reflect the N-saturated status, in which the excessive N may have reduced the productivity in N-rich systems, e.g., tropical forest (Asner et al. 2001, Fang et al. 2008). The primary stimulation was still overwhelming at regional scale, even compared to the adverse effects of N addition in recent years. We found that the C dynamics in needleleaf forests are more stimulated by enhanced N input than in broadleaf forests. The 15-year N-addition experiments in Harvard Forest also agrees with this, showing N stress in needleleaf forest tends to be relieved at a faster pace and the subsequent N saturation leads to a reduction in productivity while NPP is still increasing in broadleaf forest with the same N addition level (Magill et al. 2004).

Interactive effects of N deposition and other environmental factors

Our estimates of N-induced C sink include not only the direct contribution of atmospheric N input, but also its interactive effects with other environmental factors. Model simulations show that there exists large disagreement between the estimations of N effects with and without considering the interactive impacts of multiple driving forces. We find that CO_2 fertilization has accelerated N-induced C sequestration in most ecosystems. Model simulation indicates that N-induced C sink is reduced to 53 Tg C/yr or decreased by 15% with CO_2 fertilization effect turned off and the accumulated C sequestration is lowered to 5.62 Pg C over the past 105 years as well. A variety of FACE (free-air CO_2 enrichment) experiments with multiple N input levels conducted in China (Yang et al. 2006) and other countries (Oren et al. 2001, Kim et al. 2003, Reich et al. 2006*a*) provide evidence that elevated CO_2 can yield a larger plant growth response to N addition. Although these manipulated experiments also suggest a decline of plant growth at elevated CO_2 levels or high N supply, or both, due to the counteractive effects from excessive N or N depletion caused by CO_2 enrichment, this phenomenon has not been found in regional model simulations driven by ambient CO_2 concentration and N deposition. It is commonly accepted that elevated CO_2 could increase plant biomass, net primary production, and plant demand for N, and our model simulations also conclude that the coupling effects of $CO_2 \times N$ are larger than the sum of CO_2 and N enrichment effects alone (Tian et al. 2011*b*). In addition, for a given N input level, field experiments discovered that elevated CO_2 increased N use efficiency and widened ecosystem C:N ratios (Finzi et al. 2006), which could be two important mechanisms responsible for the delaying onset of progressive N limitation (Luo et al. 2004).

O_3 pollution and excessive N fertilization have cut down the growth response to enhanced N deposition from the atmosphere due to two different reasons: (1) plant exposure to O_3 pollution will lower its photosynthetic capability and the consequent response to N enrichment (Ren et al. 2007, 2010*a*); (2) excessive N fertilizer uses in cropland will eliminate N deficit of plant growth, and therefore decrease the response of net carbon storage to atmospheric N input (e.g., Fig. 2a shows that biomass increment peaked in the low N

fertilization level and then declined with higher doses of N input). Changes in climate and land use types have imposed complicated influence and compensated by themselves in the long run. In total, the C storage stimulated by altered N deposition alone sums up to 6.65 Pg C over the past 105 years, which is 140 Tg C larger than the estimated C storage with consideration of the interactive effects. So far, it's still difficult to verify the magnitude of such interactions at regional scale due to the lack of multifactor field experiments. The reported responses in the face of interactive stresses are deviated or even controversial, and no general patterns are ubiquitous owing to the bias introduced by various local conditions (Kim et al. 2003, Reich et al. 2006*b*, Yang et al. 2006). Long-term manipulated experiments are anticipated to involve the interactions of multiple environmental factors and the underlying mechanisms need to be extrapolated to region and globe through model development.

Uncertainty and future research needs

On regional and global scales, the simulation discrepancies among model studies were mainly resulted from the disagreement on the following three aspects: (1) The consideration of the spatial distribution and magnitude of atmospheric N deposition (Townsend et al. 1996, Holland et al. 1997). (2) The assumption and representation of biogeochemical processes in ecosystem models, e.g., how much N is retained in soils with a much lower C:N ratio (10–30) while the rest enters into the trees with higher C:N ratio (Nadelhoffer et al. 1999)? Does the model incorporate C–N coupling as a feedback in order to avoid overestimation on CO_2 or N fertilization effects (Hungate et al. 2003, Sokolov et al. 2008, Jain et al. 2009, Zaehle et al. 2010*a*, *b*)? Is the N cycle in the model fully open and allowed to exchange with exterior environment (Rastetter et al. 1997)? (3) The inclusion of interaction among N deposition and changes in climate, atmospheric composition (CO_2, O_3), land use history, and land management practices in the simulation (Churkina et al. 2007). In the following, we discuss the uncertainties caused by these three aspects.

We conducted the same three simulations using the modeled N deposition data from Dentener (2006), which has been widely used for assessing the N effects on terrestrial ecosystems. We find that the ecosystem C uptake driven by modeled N input was far less than the value estimated by field observation-derived N deposition pattern used in this study (Lu and Tian 2007). Simulation driven by modeled N deposition data show that the N-induced C sink averaged 34 Tg C/yr, and added up to 3.61 Pg C during 1901–2005, accounting for only 56% of the C sink as simulated using our field observation-based N deposition data. Obviously, the uncertainty of estimating N-induced C sequestration is highly associated with N deposition amount, spatial and temporal distribution. Although N input data we used in this study is close to levels observed in the field, the inherent uncertainties resulting from site representativeness, data sampling and interpolation approach (Lu and Tian et al. 2007) have undoubtedly biased our estimation through DLEM simulation. Moreover, the lack of information on the seasonal N deposition pattern and various deposited N species (e.g., organic N) monitoring also has influenced the simulated N impacts. Sufficient research to monitor N deposition from the atmosphere over a long period is urgently needed for obtaining more reliable input data.

In addition, uncertainty derived by model structure and assumptions cannot be ignored in estimating N deposition effects on the terrestrial ecosystems' C dynamics, which is likely to contribute to the large discrepancies among different estimations (Hungate et al. 2003, Sokolov et al. 2008). Ecosystem models have distinct strategies in partitioning and describing the pools/fluxes and C–N interactions, as well as in simulating N-saturated status. Whether the model's representation of C–N coupling can truly reflect the C and N cycling in the real world is a hard nut to crack for most models involving C–N interactions. As Zaehle et al. (2010*a*) pointed out, even though all the models agreed that N limitation will reduce the CO_2 fertilization effect and warming-enhanced mineralization can increase N availability and therefore stimulate plant growth, they would not give the same significance to these two counteractive effects. The divergence on the estimated magnitude of various C–N coupling processes is a major reason responsible for the contrasting results among modeling studies (Sokolov et al. 2008, Jain et al. 2009, Thornton et al. 2009, Zaehle et al. 2010*a*). Therefore, field experiments focusing on C and N processes and data–model intercomparison programs, such as LBA-DMIP (Large Scale Biosphere–Atmosphere Experiment of the Amazon, Data Model Intercomparison Project) and NACP (North America Carbon Program), are highly needed to diminish this divergence in model development. Intercomparison of various models with the same input data is also expected to identify the critical processes, parameters and assumptions in determining the N-stimulated C uptake. As for DLEM, the model assumption and representation of C–N coupling have been evaluated through the following aspects: (1) the modeled responses of productivity in several major biome types fall in the range of experimental results; (2) the sensitivity of biomass and nitrogen leaching loss in response to different doses of N addition simulated by DLEM is consistent with the observations and research findings; (3) the simulated N saturation signs are also confirmed by the experimental evidence.

In this study, we include cropland expansion, and natural vegetation colonization and development on abandoned cropland, to depict land use and cover changes. Forest recovery is included when new forests start to grow after cropland abandonment and natural and anthropogenic disturbances (fire, clear cuts, hurri-

canes, and so on), but we do not consider the role of age structure-related forest regrowth in estimating C dynamics in response to N enrichment. Churkina et al. (2007) suggested that regrowing forests would amplify the impacts of increased N deposition relative to mature forests. Field experiments in tropical forests of southern China also showed that the biomass production of seedlings in response to N addition could be increased by 110% relative to the control level (Mo et al. 2008), far beyond the growth responses of mature forests. However, we simplify the representation of ecosystem structure, such as species composition, phenology, maximum biomass, and mortality, due to the lack of detailed records of plantation, disturbance history, and ecosystem succession. This, to some extent, underestimates the forest C sink induced by enhanced N deposition because reforestation and afforestation has occurred in large areas of China since the 1950s (Liu and Tian 2010). Forests of different age classes and contrasting growth capability ought to be incorporated into the future modeling work for N impacts estimation. In addition, agronomic practices, such as crop cultivar improvement, irrigation, rotation, and residual return, have been simply considered in order to implement DLEM's simulation across cropland (Ren et al. 2011*b*), but the further explicit analysis requests long-term historical records to describe the distribution, frequency, and magnitude of these agricultural activities and their impacts on crop response to N enrichment.

Conclusion

Our simulated results indicate that atmospheric N deposition caused a net C sink of 62 Tg C/yr, adding up to 6.51 Pg C on a national scale for the period 1901–2005. N-induced C sequestration demonstrates an increasing gradient from the northern and northwestern China to the southeastern China, which corresponds to an increasing gradient of N deposition levels. Divergent patterns of biome-level response to increased N deposition exist due to inherently different mechanisms and co-limitation from other environmental factors. Since the 1980s, ecosystem N use efficiency has begun to level off or decline, indicating soil N retention capability has reached its peak, and the additional N may not further stimulate C uptake, but instead leave the ecosystem through leaching and nitrous gas emissions. In large areas and major ecosystem types of China, signs of N saturation have been apparently revealed in recent decades. Hence, although our study indicates that atmospheric N deposition has enhanced C uptake of terrestrial ecosystems in China over the past century, the potentially detrimental effect of excessive N input, such as soil degradation, water acidification and accelerated emission of N_2O, a potent greenhouse gas, should be paid more attention in the near future. In order to increase terrestrial C sink in China, effective management practices should be taken to improve N use efficiency rather than to keep N input level continuing to rise.

To accurately assess the N deposition impact on C sequestration in terrestrial ecosystems, it is important to take into account the interactive effects of N deposition with other environmental changes. Our simulation suggests that the overall interaction between N deposition and other factors (climate, CO_2, O_3, land use, N fertilizer application) reduced the C sink due to increasing N deposition alone by 140 Tg C for the study period. Elevated CO_2 concentration is among the most contributing factors, and the N-induced C storage would shrink to 5.62 Pg C if the CO_2 fertilization effect was shut off. In addition, O_3 pollution and excessive N fertilization have cut down the growth response to atmospheric N deposition. Changes in climate and land use types have imposed complicated influence and varied over time and space.

To the best of our knowledge, this work is the very first estimation of N deposition impacts on C balance in China's terrestrial ecosystems over the past century. To reduce uncertainties in estimating effects of increasing N deposition on terrestrial C sequestration and C storage, however, future work needs to develop more reliable N deposition data, and to focus on more accurate estimates on processes relevant to N budget and ecosystem retentions. The chronic low-dose N addition experiments are important to improve model representation of ecosystem responses to increased N deposited from the atmosphere. To better simulate the interactive effects of N deposition with other environmental factors, we call for field manipulative experiments including multiple environmental factors for providing modeling work with a solid knowledge of the interactions between N addition and the changes in supply of heat, water, carbon and other nutrient elements. Clearly, the rigorous interaction between modeling and field experimentation will be essential to enhance our understanding of C–N coupling in terrestrial ecosystems in the context of multifactor environmental changes.

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Literature Cited

Aber, J. D., W. McDowell, K. Nadelhoffer, A. Magill, G. Berntson, M. Kamakea, S. McNulty, W. Currie, L. Rustad, and I. Fernandez. 1998. Nitrogen saturation in temperate forest ecosystems: hypotheses revisited. BioScience 48:921–934.

Ainsworth, E. A., and S. P. Long. 2005. What have we learned from 15 years of free-air CO_2 enrichment (FACE)? A meta-analytic review of the responses of photosynthesis, canopy. New Phytologist 165:351–371.

Asner, G. P., A. R. Townsend, W. J. Riley, P. A. Matson, J. C. Neff, and C. C. Cleveland. 2001. Modeling tropical and semi-arid ecosystem responses to increased N deposition. Biogeochemistry 54:1–39.

Boden, T. A., G. Marland, and R. J. Andres. 2009. Global, regional, and national fossil-fuel CO_2 emissions. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tennessee, USA. http://dx.doi.org/10.3334/CDIAC/00001

Bowden, R. D., E. Davidson, K. Savage, C. Arabia, and P. Steudler. 2004. Chronic nitrogen additions reduce total soil respiration and microbial respiration in temperate forest soils at the Harvard Forest. Forest Ecology and Management 196:43–56.

Chen, G., H. Tian, M. Liu, W. Ren, C. Zhang, and S. Pan. 2006. Climate impacts on China's terrestrial carbon cycle: an assessment with the dynamic land ecosystem model. Pages 56–70 *in* H. Q. Tian, editor. Environmental modeling and simulation. ACTA Press, Calgary, Alberta, Canada.

Churkina, G., K. Trusilova, M. Vetter, and F. Dentener. 2007. Contribution of nitrogen deposition and forest regrowth to terrestrial carbon uptake. Carbon Balance and Management 2:5.

Dentener, F. J. 2006. Global maps of atmospheric nitrogen deposition, 1860, 1993, and 2050. Data set. Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, Tennessee, USA.

De Vries, W., S. Solberg, M. Dobbertin, H. Sterba, D. Laubhann, M. van Oijen, C. Evans, P. Gundersen, J. Kros, G. W. W. Wamelink, G. J. Reinds, and M. A. Sutton. 2009. The impacts of nitrogen deposition on carbon sequestration by European forest and heathlands. Forest Ecology and Management 258:1814–1823.

Elser, J. J., M. E. S. Bracken, E. E. Cleland, D. S. Gruner, W. S. Harpole, H. Hillebrand, J. T. Ngai, E. W. Seabloom, J. B. Shurin, and J. E. Smith. 2007. Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine, and terrestrial ecosystems. Ecology Letters 10:1135–1142.

Fang, Y. T., P. Gundersen, J. M. Mo, and W. X. Zhu. 2008. Input and output of dissolved organic and inorganic nitrogen in subtropical forests of South China under high air pollution. Biogeosciences 5:339–352.

Fang, Y. T., P. Gundersen, J. M. Mo, and W. X. Zhu. 2009. Nitrogen leaching in response to increased nitrogen inputs in subtropical monsoon forests in southern China. Forest Ecology and Management 257:332–342.

Felzer, B. S., J. Reilly, J. Melillo, D. W. Kicklighter, M. Sarofim, C. Wang, R. G. Prinn, and Q. Zhuang. 2005. Future effects of ozone on carbon sequestration and climate change policy using a global biochemistry model. Climate Change 73:345–373.

Fenn, M. E., J. S. Baron, E. B. Allen, H. M. Rueth, K. R. Nydick, L. Geiser, W. D. Bowman, J. O. Sickman, T. Meixner, D. W. Johnson, and P. Neitlich. 2003. Ecological effects of nitrogen deposition in the western United States. BioScience 53:404–419.

Finzi, A. C., D. J. Moore, E. H. DeLucia, J. Lichter, K. S. Hofmockel, R. B. Jackson, H. Kim, R. Matamala, H. R. McCarthy, R. Oren, J. S. Pippen, and W. H. Schlesinger. 2006. Progressive nitrogen limitation of ecosystem processes under elevated CO_2 in a warm-temperate forest. Ecology 87:15–25.

Fisher, L. S., P. A. Mays, and C. L. Wylie. 2007. An overview of nitrogen critical loads for policy makers, stakeholders, and industries in the United States. Water, Air, Soil Pollution 179:3–18.

Galloway, J. N., et al. 2004. Nitrogen cycles: past, present, and future. Biogeochemistry 70(2):153–226.

Galloway, J. N., F. J. Dentener, E. Marmer, Z. Cai, Y. P. Abrol, V. K. Dahwal, and A. V. Murugan. 2008*a*. The environmental reach of Asia. Annual Review of Environment and Resources 33:461–481.

Galloway, J. N., A. R. Townsend, J. W. Erisman, M. Bekunda, Z. Cai, J. R. Freney, L. A. Martinelli, S. P. Seitzinger, and M. A. Sutton. 2008*b*. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. Science 320(5878):889–892.

Gifford, R. M., J. L. Lutze, and D. Barrett. 1996. Global atmospheric change effects on terrestrial carbon sequestration: exploration with a global C- and N-cycle model (CQUESTN). Plant and Soil 187:369–387.

Högberg, P., H. Fan, M. Quist, D. Binkley, and C. O. Tamm. 2006. Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest. Global Change Biology 12:489–499.

Holland, E. A., B. H. Braswell, J. F. Lamarque, A. Townsend, J. Sulzman, J.-F. Müller, F. Dentener, G. Brasseur, H. Levy II, J. E. Penner, and G.-J. Roelofs. 1997. Variations in the predicted spatial distribution of atmospheric nitrogen deposition and their impact on carbon uptake by terrestrial ecosystems. Journal of Geophysical Research 102:849–866.

Holland, E. A., B. H. Braswell, J. Sulzman, and J. F. Lamarque. 2005. Nitrogen deposition onto the United States and Western Europe: synthesis of observation and models. Ecological Application 15:38–57.

Hungate, B. A., J. S. Dukes, M. R. Shaw, Y. Q. Luo, and C. B. Field. 2003. Nitrogen and climate change. Science 302:1512.

Jain, A., X. Yang, H. Kheshgi, A. D. McGuire, W. Post, and D. Kicklighter. 2009. Nitrogen attenuation of terrestrial carbon cycle response to global environmental factors. Global Biogeochemical Cycles. http://dx.doi.org/10.1029/2009GB003519

Ju, X. T., G. Xing, X. Chen, S. Zhang, L. Zhang, X. Liu, Z. Cui, B. Yin, P. Christie, Z. Zhu and F. Zhang. 2009. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. Proceedings of the National Academy of Sciences USA 106:3041–3046.

Kim, H. Y., M. Lieffering, K. Kobayashi, M. Okada, M. W. Mitchell, and M. Gumpertz. 2003. Effects of free-air CO_2 enrichment and nitrogen supply on the yield of temperate paddy rice crops. Field Crops Research 83:261–270.

LeBauer, D. S., and K. K. Treseder. 2008. Nitrogen limitation of net primary productivity in terrestrial ecosystems is globally distributed. Ecology 89:371–379.

Liu, J. Y., M. L. Liu, H. Q. Tian, D. Zhuang, Z. Zhang, W. Zhang, X. Tang, and X. Deng. 2005*a*. Current status and recent changes of cropland in China: an analysis based on Landsat TM data. Remote Sensing and Environment 98:442–456.

Liu, J. Y., H. Q. Tian, M. L. Liu, D. Zhuang, J. M. Melillo, and Z. Zhang. 2005*b*. China's changing landscape during the 1990s: large-scale land transformation estimated with satellite data. Geophysical Research Letters 32:L02405.

Liu, M. L., and H. Q. Tian. 2010. China's land-cover and land-use change from 1700 to 2005: estimations from high-resolution satellite data and historical archives. Global Biogeochemical Cycles 24:GB3003.

Liu, M. L., H. Q. Tian, G. S. Chen, W. Ren, C. Zhang, and J. Liu. 2008. Effects of land use and land cover change on evapotranspiration and water yield in China during the 20th century. Journal of the American Water Resources Association 44(5):1193–1207.

Lu, C. Q. 2009. Atmospheric nitrogen deposition and terrestrial ecosystem carbon cycle in China. Dissertation. Chinese Academy of Sciences, Beijing, China.

Lu, C. Q., and H. Q. Tian. 2007. Spatial and temporal patterns of nitrogen deposition in China: synthesis of observational data. Journal of Geophysical Research 112:D22S05.

Lu, C. Q., H. Q. Tian, and Y. Huang. 2007. Ecological effects of increasing nitrogen deposition in terrestrial ecosystems. Journal of Plant Ecology 31:205–218.

Lu, F., X. Wang, B. Han, Z. Quyang, X. Duan, H. Zheng, and H. Miao. 2009. Soil carbon sequestrations by nitrogen fertilizer application, straw return and no-tillage in China's cropland. Global Change Biology 15:281–305.

Luo, Y., et al. 2004. Progressive nitrogen limitation of ecosystem responses to rising atmospheric CO_2. BioScience 54:731–739.

Magill, A. H., J. D. Aber, G. M. Bertson, W. H. McDowell, K. J. Nadelhoffer, J. M. Melillo, and P. Steudler. 2000. Long-term nitrogen additions and nitrogen saturation in two temperate forests. Ecosystems 3:238–253.

Magill, A. H., J. D. Aber, W. S. Currie, K. J. Nadelhoffer, M. E. Martin, W. H. McDowell, J. M. Melillo and P. Steudler. 2004. Ecosystem response to 15 years of chronic nitrogen additions at the Harvard Forest LTER, Massachusetts, USA. Forest Ecology and Management 196:7–28.

Magnani, F., et al. 2007. The human footprint in the carbon cycle of temperate and boreal forests. Nature 447:848–850.

Matson, P. A., W. H. McDowell, A. R. Townsend, and P. M. Vitousek. 1999. The globalization of N deposition: ecosystem consequences in tropical environments. Biogeochemistry 46:67–83.

Mo, J. M., D. J. Li, and P. Gundersen. 2008. Seedling growth response of two tropical tree species to nitrogen deposition in southern China. European Journal of Forest Research 127:125–283.

Mo, J. M., J. H. Xue, and Y. T. Fang. 2004. Litter decomposition and its responses to simulated N deposition for the major plants of Dinghushan forests in subtropical China. Acta Ecologica Sinica 24:1413–1420.

Nadelhoffer, K. J., B. P. Colman, W. S. Currie, A. H. Magill, and J. D. Aber. 2004. Decadal scale fates of ^{15}N tracers added to oak and pine stands under ambient and elevated N inputs at the Harvard Forest (USA). Forest Ecology and Management 196:89–107.

Nadelhoffer, K. J., B. A. Emmet, P. Gundersen, O. J. Kjønaas, C. J. Koopmans, P. Schleppi, A. Tietema, and R. F. Wright. 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. Nature 398:145–148.

Niu, S., M. Wu, Y. Han, J. Xia, Z. Zhang, H. Yang, and S. Wan. 2009. Nitrogen effects on net ecosystem carbon exchange in a temperate steppe. Global Change Biology. http://dx.doi.org/10.1111/j.1365-2486.2009.01894.x

Oren, R., D. Ellsworth, K. Johnsen, N. Phillips, B. E. Ewers, C. Maier, K. V. R. Schäfer, H. McCarthy, G. Hendrey, S. G. McNulty, and G. G. Katul. 2001. Soil fertility limits carbon sequestration by forest ecosystems in a CO2-enriched atmosphere. Nature 411:469–472.

Rastetter, E. B., G. I. Agren, and G. R. Shaver. 1997. Responses of N-limited ecosystems to increased CO_2: a balanced-nutrition, coupled-element-cycles model. Ecological Application 7:444–460.

Reich, P. B., S. E. Hobbie, T. Lee, D. S. Ellsworth, J. B. West, D. Tilman, J. M. H. Knops, S. Naeem, and J. Trost. 2006*a*. Nitrogen limitation constrains sustainability of ecosystem response to CO_2. Nature 440:922–925.

Reich, P. B., B. A. Hungate, and Y. Luo. 2006*b*. Carbon–nitrogen interactions in terrestrial ecosystems in response to rising atmospheric CO_2. Annual Review of Ecology, Evolution, and Systematics 37:611–636.

Ren, W., H. Tian, M. Liu, C. Zhang, G. Chen, S. Pan, B. Felzer, and X. Xu. 2007. Effects of tropospheric ozone pollution on net primary productivity and carbon storage in terrestrial ecosystems of China. Journal of Geophysical Research 112:D22S09.

Ren, W., H. Tian, B. Tao, A. Chappelka, G. Sun, C. Lu, M. Liu, G. Chen, and X. Xu. 2011*a*. Impacts of ozone pollution and climate change on net primary productivity and carbon storage of China's forest ecosystems as assessed by using a process-based ecosystem model. Global Ecology and Biogeography. http://dx.doi.org/10.1111/j.1466-8283.2010.00606.x

Ren, W., H. Q. Tian, X. F. Xu, M. L. Liu, C. Q. Lu, G. S. Chen, J. Mellio, J. Reilly, and J. Y. Liu. 2011*b*. Spatial and temporal patterns of CO_2 and CH_4 fluxes in China's croplands in response to multifactor environmental changes. Tellus B. http://dx.doi.org/10.1111/j.1600-0889.2010.00522.x

Richter, A., J. P. Burrows, H. Nüß, C. Granier, and U. Niemeier. 2005. Increase in tropospheric nitrogen dioxide over China observed from space. Nature 437:129–132.

Schindler, D. W., and S. E. Bayley. 1993. The biosphere as an increasing sink for atmospheric carbon: Estimates from increased nitrogen deposition. Global Biogeochemical Cycles 7:717–733.

Schwalm, C. R., C. A. Williams, K. Schaefer, R. Anderson, M. A. Arain, I. Baker, T. A. Black, G. Chen, P. Ciais, K. J. Davis, and other NACP participants. 2010. A model-data intercomparison of CO_2 exchange across North America: Results from the North American Carbon Program Site Synthesis. Journal of Geophysical Research. http://dx.doi.org/10.1029/2009JG001229

Shi, X. Z., D. S. Yu, E. D. Warner, X. Z. Pan, G. W. Petersen, Z. G. Gong, and W. G. Weindorf. 2004. Soil database of 1:1,000,000 digital soil survey and reference system of the Chinese genetic soil classification system. Soil Survey Horizons 45:139–136.

Shui, J., X. Chai, and T. Lu. 1999. Effects of precipitation on nutrient inputs and erosion of forest land. Soil and Environmental Sciences 8(4):258–261. [In Chinese with English abstract.]

Sokolov, A. P., D. W. Kicklighter, J. M. Melillo, B. S. Felzer, C. A. Schlosser, and T. W. Cronin. 2008. Consequences of considering carbon–nitrogen interactions on the feedbacks between climate and the terrestrial carbon cycle. Journal of Climate 21:3776–3796.

Thornton, P. E., S. C. Doney, K. Lindsay, J. K. Moore, N. Mahowald, J. T. Randerson, I. Fung, J.-F. Lamarque, J. J. Feddema, and Y.-H. Lee. 2009. Carbon–nitrogen interactions regulate climate-carbon cycle feedbacks: results from an atmospheric-ocean general circulation model. Biogeosciences 6:2099–2120.

Tian, H. Q., G. S. Chen, M. L. Liu, C. Zhang, G. Sun, C. Lu, X. Xu, W. Ren, S. Pan, and A. Chappelka. 2010*a*. Model estimates of ecosystem net primary productivity, evapotranspiration, and water use efficiency in the southern United States during 1895–2007. Forest Ecology and Management 259:1311–1327.

Tian, H. Q., G. S. Chen, C. Zhang, J. M. Melillo, and C. Hall. 2010*b*. Patterns and variations of C:N:P ratios in China's soils: a synthesis of observational data. Biogeochemistry. http://dx.doi.org/10.1007/s10533-009-9382-0

Tian, H. Q., M. Liu, C. Zhang, W. Ren, X. Xu, G. Chen, C. Lu, and B. Tao. 2010*c*. The dynamic land ecosystem model (DLEM) for simulating terrestrial processes and interactions in the context of multifactor global change. Acta Geographical Science 65(9):1027–1047.

Tian, H. Q., C. Q. Lu, G. S. Chen, X. F. Xu, M. L. Liu, W. Ren, B. Tao, G. Sun, S. F. Pan, and J. Y. Liu. 2011*a*. Controls of climate and land use over terrestrial water use efficiency in monsoon Asia during the late half of 20th Century. Ecohydrology 4:322–340.

Tian, H. Q., et al. 2011*b*. China's terrestrial carbon balance: Contributions from multiple global change factors. Global Biogeochemical Cycles. http://dx.doi.org/10.1029/2010GB003838

Tian, H. Q., S. Wang, J. Liu, S. Pan, H. Chen, C. Zhang, and X. Shi. 2006. Patterns of soil nitrogen storage in China. Global Biogeochemical Cycle 20:GB1001.

Tian, H. Q., X. F. Xu, M. L. Liu, W. Ren, C. Zhang, G. Chen, and C. Lu. 2010*d*. Spatial and temporal patterns of CH_4 and N_2O fluxes in terrestrial ecosystems of North America during

1979-2008: application of a global biogeochemistry model. Biogeosciences 7:2673–2694.

Tian, H. Q., X. F. Xu, C. Q. Lu, M. L. Liu, W. Ren, G. S. Chen, J. Melillo, and J. Y. Liu. 2011*c*. Net exchanges of CO_2, CH_4, and N_2O between China's terrestrial ecosystems and the atmosphere and their contributions to global climate warming. Journal of Geophysical Research. http://dx.doi.org/10.1029/2010JG001393

Tian, H. Q., X. Xu, C. Zhang, W. Ren, G. Chen, M. Liu, D. Lu, S. Pan. 2008. Forecasting and assessing the large-scale and long-term impacts of global environmental change on terrestrial ecosystems in the United States and China. *In* S. Miao, S. Carstenn, and M. Nungesser, editors. Real world ecology: large-scale and long-term case studies and methods. Springer-Verlag, New York, New York, USA.

Tietema, A., B. A. Emmett, P. Gundersen, O. J. Kjønaas and C. J. Koopmans. 1998. The fate of ^{15}N-labelled nitrogen deposition in coniferous forest ecosystem. Forest Ecology and Management 101:19–27.

Townsend, A. R., B. H. Braswell, E. A. Holland, and J. E. Penner. 1996. Spatial and temporal patterns in terrestrial carbon storage due to deposition of anthropogenic nitrogen. Ecological Applications 6:806–814.

Vitousek, P. M., J. D. Aber, R. W. Howarth, G. E. Likens, P. A. Matson, D. W. Schindler, W. H. Schlesinger, and D. G. Tilman. 1997. Human alteration of the global nitrogen cycle: Sources and consequences. Ecological Applications 7:737–750.

Vitousek, P. M., and R. W. Howarth. 1991. Nitrogen limitation on land and in the sea: How can it occur? Biogeochemistry 13:87–115.

Wang, X. W., Q. Yu, G. L. Zhang, and Y. S. Li. 2005. The yield and dynamics of soil nitrate for summer maize on the plain of northwest Shandong province. Geographical Research 24:140–150.

Wright, R. F., and L. Rasmussen. 1998. Introduction to the NITREX and EXMAN projects. Forest Ecology and Management 101:1–7.

Xia, J. Y., and S. Q. Wan. 2008. Global response patterns of terrestrial plant species to nitrogen addition. New Phytologist 179:428–439.

Xu, X., H. Tian, C. Zhang, M. Liu, W. Ren, G. Chen, and C. Lu. 2010. Attribution of spatial and temporal variations in terrestrial methane flux over North America. Biogeosciences 7:3637–3655.

Xu-Ri, and I. C. Prentice. 2008. Terrestrial nitrogen cycle simulation with a dynamic global vegetation model. Global Change Biology 14: 1745–1764.

Yang, L. X., J. Y. Huang, H. J. Yang, J. Zhu, H. Liu, G. Dong, G. Liu, Y. Han, and Y. Wang. 2006. The impact of free-air CO_2 enrichment (FACE) and N supply on yield formation of rice crops with large panicle. Field Crops Research 98:141–150.

Zaehle, S., P. Friedlingstein, and A. D. Friend. 2010*a*. Terrestrial nitrogen feedbacks may accelerate future climate change. Geophysical Research Letters. http://dx.doi.org/10.1029/2009GL041345

Zaehle, S., A. D. Friend, P. Friedlingstein, F. Dentener, P. Peylin, and M. Schulz. 2010*a*. Carbon and nitrogen cycle dynamics in the O-CN land surface model: 2. Role of the nitrogen cycle in the historical terrestrial carbon balance. Global Biogeochemical Cycles 24:GB1006.

Zhang, C., H. Q. Tian, A. Chappelka, W. Ren, H. Chen, S. Pan, M. Liu, D. M. Styers, G. Chen, and Y. Wang. 2007. Impacts of climatic and atmospheric changes on carbon dynamics in the Great Smoky Mountains National Park. Environmental Pollution 149:336–347.

Zhang, C., H. Q. Tian, Y. Wang, T. Zeng, and Y. Liu 2010. Predicting response of fuel load to future changes in climate and atmospheric composition in the Southern United States. Forest Ecology and Management 260:556–564.

Supplemental Material

Appendix

Model structure and equations related to nitrogen budget and nitrogen control on carbon processes, used in the dynamic land ecosystem model (DLEM) (*Ecological Archives* A022-003-A1).