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PAPER

# Impacts of tropospheric ozone and climate change on net primary productivity and net carbon exchange of China's forest ecosystems

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## ABSTRACT

**Aim** We investigated how ozone pollution and climate change/variability have interactively affected net primary productivity (NPP) and net carbon exchange (NCE) across China's forest ecosystem in the past half century.

**Location** Continental China.

**Methods** Using the dynamic land ecosystem model (DLEM) in conjunction with 10-km-resolution gridded historical data sets (tropospheric O<sub>3</sub> concentrations, climate variability/change, and other environmental factors such as land-cover/land-use change (LCLUC), increasing CO<sub>2</sub> and nitrogen deposition), we conducted nine simulation experiments to: (1) investigate the temporo-spatial patterns of NPP and NCE in China's forest ecosystems from 1961–2005; and (2) quantify the effects of tropospheric O<sub>3</sub> pollution alone or in combination with climate variability and other environmental stresses on forests' NPP and NCE.

**Results** China's forests acted as a carbon sink during 1961–2005 as a result of the combined effects of O<sub>3</sub>, climate, CO<sub>2</sub>, nitrogen deposition and LCLUC. However, simulated results indicated that elevated O<sub>3</sub> caused a 7.7% decrease in national carbon storage, with O<sub>3</sub>-induced reductions in NCE (Pg C year<sup>-1</sup>) ranging from 0.4–43.1% among different forest types. Sensitivity experiments showed that climate change was the dominant factor in controlling changes in temporo-spatial patterns of annual NPP. The combined negative effects of O<sub>3</sub> pollution and climate change on NPP and NCE could be largely offset by the positive fertilization effects of nitrogen deposition and CO<sub>2</sub>.

**Main conclusions** In the future, tropospheric O<sub>3</sub> should be taken into account in order to fully understand the variations of carbon sequestration capacity of forests and assess the vulnerability of forest ecosystems to climate change and air pollution. Reducing air pollution in China is likely to increase the resilience of forests to climate change. This paper offers the first estimate of how prevention of air pollution can help to increase forest productivity and carbon sequestration in China's forested ecosystems.

## Keywords

China, climate change, dynamic land ecosystem model (DLEM), forest ecosystem, net carbon exchange (NCE), net primary production (NPP), ozone (O<sub>3</sub>).

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## INTRODUCTION

In recent decades, there has been increased concern that elevated tropospheric ozone ( $O_3$ ) and climate change have influenced the ability of China's ecosystems to provide people with essential goods and services (Fu *et al.*, 2007). Between 1980 and 1995, fertilizer use in China was 36% greater than the average in developed countries, where fertilizer use has been decreasing, and 65% greater than the average in other developing countries. Both fossil fuel consumption and nitrogen fertilizer application can contribute to total emissions of  $NO_x$ , a main  $O_3$  precursor, and consequently result in increased atmospheric  $O_3$  concentrations (Chameides *et al.*, 1992). It has been estimated that China's emissions of  $NO_x$  might increase four times by the year 2020 without pollution controls, compared with 1990 emissions (van Aardenne *et al.*, 1999). This would lead to a much larger increase in tropospheric  $O_3$  with concentrations of 150 parts per billion (p.p.b.;  $\mu l^{-1}$ ) of  $O_3$  in some locations (Elliott *et al.*, 1997), particularly in southern China where  $NO_x$  emissions are increasing due to high nitrogen input derived from deposition and fertilizer application (Lu & Tian, 2007). In addition, significant climate warming trends have occurred in China in the past two decades. The 1990s were one of the warmest periods in the last 100 years, with large temporal and spatial variations in temperature and precipitation (IPCC, 2007). However, it is far from certain to what extent these changes in tropospheric  $O_3$  and climate have affected regional forest productivity and carbon sequestration in China.

Forest ecosystems play a dominant role in the terrestrial carbon (C) budget because of the large amount of C stored in vegetation and the soil (Goodale *et al.*, 2002). Forest productivity, governed by both natural factors (e.g. climate, succession and disturbance) and human activities, is regarded as a key indicator of changes in forest ecosystem structure and functioning. Recent negotiations regarding the Kyoto Protocol to the UN Framework Convention on Climate Change (UNFCCC) have focused considerable attention on forest in the context of climate change, due to its critical role in the global carbon cycle and climate system (Bonan, 2008). More than 50% of the carbon in terrestrial vegetation is stored in forest ecosystems (Dixon *et al.*, 1994). Additionally, forests could significantly affect the course of global warming in the 21st century by influencing exchanges of energy, water and greenhouse gases between the atmosphere and forests (Bonan, 2008). In China, approximately 18% of the land area is covered by forests as reported in the 6th National Forest Resources Inventory, 1999–2003 (Xiao, 2005); these forests play an important role in sequestering carbon, regulating local climate and providing other ecosystem services and goods. Most of the forested areas are distributed in the monsoon climate zone with high  $O_3$  pollution occurring in some areas.

Many previous studies have estimated vegetation/soil carbon stocks and fluxes in China's forests using different approaches, including inventory-based methods (e.g. Luo *et al.*, 1999; Fang *et al.*, 2007), process-based models (Xiao *et al.*, 1998; Tao *et al.*, 2007), and remote sensing-based methods (e.g. Jiang *et al.*,

1999) at different temporo-spatial scales. Most studies indicated that net primary productivity (NPP) in China's forest ecosystems, including plantations and natural forests, has increased since the 1970s. These forest ecosystems acted as carbon sources of  $0.013 \text{ Pg C year}^{-1}$  (Fang *et al.*, 2001) or carbon sinks of  $0.020 \text{ Pg C year}^{-1}$  between 1977 and 1981 (Pan *et al.*, 2004), and then became carbon sinks with increasing carbon sequestration occurring during the 1980s and the 1990s (Fang *et al.*, 2001, 2007; Pan *et al.*, 2004; Tao *et al.*, 2007; Zhuang *et al.*, 2009). Although process-based modelling studies have focused on the forest carbon cycle in response to climate change (Xiao *et al.*, 1998; Tao *et al.*, 2007), little is known about how tropospheric  $O_3$  concentrations have influenced the growth and productivity of China's forest ecosystems. Quantifying the impact of current and future effects of  $O_3$  on forests is an urgent task (e.g. Wittig *et al.*, 2009). The spatially explicit ecosystem model is a useful tool for assessing the regional impacts of climate change and other environmental stresses, but only a few existing models that estimate the carbon sequestration potential of forest ecosystems consider  $O_3$  pollution (e.g. Ollinger *et al.*, 1997, 2002; Felzer *et al.*, 2004, 2005; Sitch *et al.*, 2007), and little research has been conducted addressing the impacts of  $O_3$  in combination with climate change on NPP and net carbon exchange (NCE) across China's forest ecosystems.

In this study we investigated the potential effects of elevated  $O_3$  along with climate change/variability on NPP and NCE in China's forest ecosystems for the period 1961–2005 using a process-based dynamic land ecosystem model (DLEM; Tian *et al.*, 2005, 2010a,b). In addition to the historical information regarding  $O_3$  pollution and climate change, we considered other major environmental factors as model inputs, including atmosphere  $CO_2$ , nitrogen deposition, land-use change and tree regrowth. The objectives of our study were to: (1) quantify the effects of tropospheric  $O_3$  pollution in combination with climate variability on NPP and NCE of China's forest ecosystems from 1961–2005; (2) examine the temporo-spatial patterns of NPP and NCE in China's forest ecosystems during 1961–2005; (3) investigate the varied sensitivities of different forest types in response to  $O_3$  pollution; (4) attribute the interactive effects of  $O_3$  and climate combined with other environmental factors (nitrogen deposition,  $CO_2$ ) on forest productivity and the rate of carbon sequestration.

## MATERIALS AND METHODS

### The dynamic land ecosystem model (DLEM)

The DLEM is a process-based model that couples major biogeochemical cycles, hydrological processes and vegetation dynamics to generate daily, spatially explicit estimates of water, carbon ( $CO_2$ ,  $CH_4$ ) and nitrogen fluxes ( $N_2O$ ,  $NH_3$ ,  $NH_4$ ,  $NO_x$ ) and pool sizes (C and N) in terrestrial ecosystems (Tian *et al.*, 2005, 2010a,c; Ren *et al.*, 2007a,b; Liu *et al.*, 2008; see Fig. 1). The DLEM includes five core components: (1) biophysics, (2) plant physiology, (3) soil biogeochemistry, (4) dynamic vegetation, and (5) disturbances, land use and management. The bio-

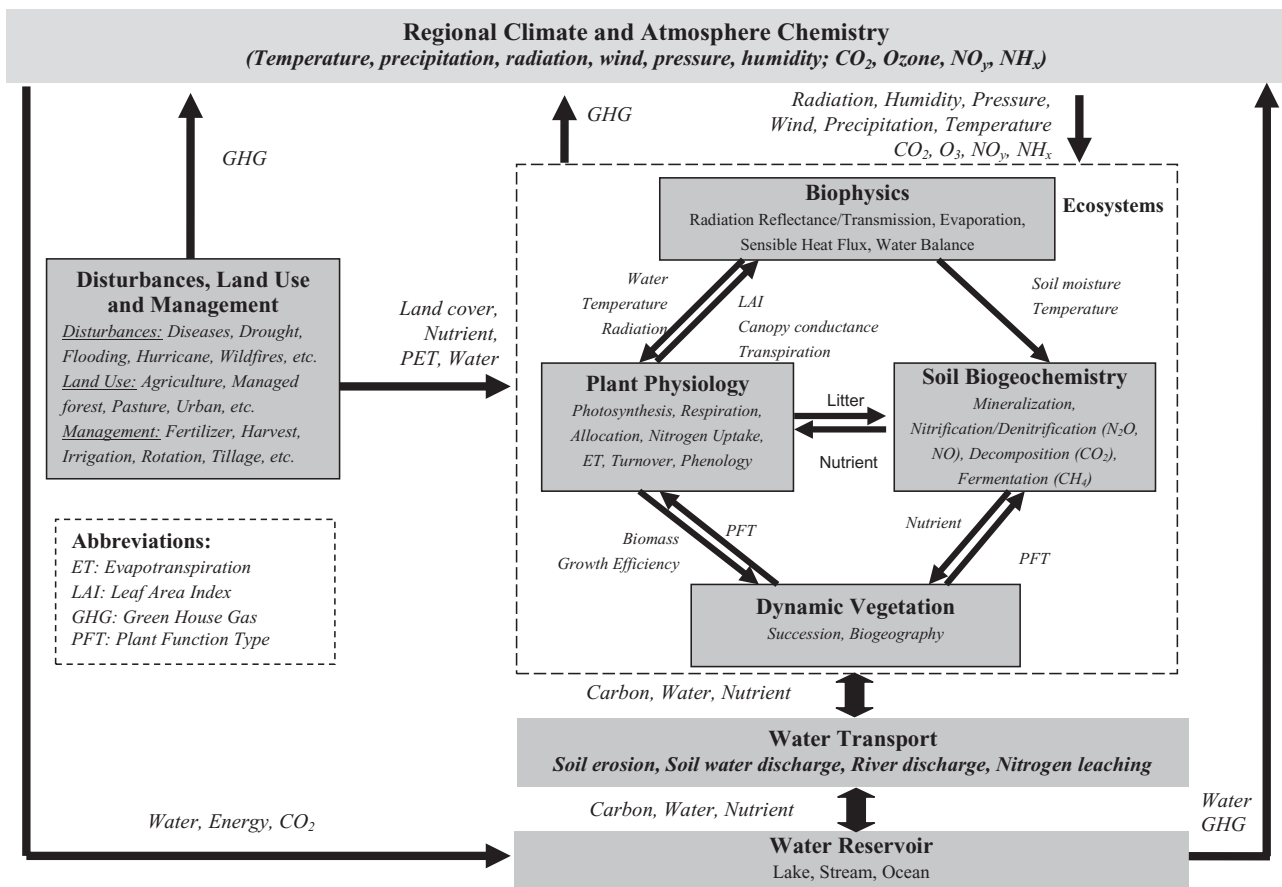


Figure 1 Framework of the dynamic land ecosystem model (DLEM) (Tian *et al.*, 2005, 2010a,c).

physical component includes the instantaneous exchanges of energy, water and momentum with the atmosphere. The model also includes aspects of micrometeorology, canopy physiology, soil physics, radiative transfer, hydrology, surface fluxes of energy, moisture, and influences of momentum on simulated surface climate. The component of plant physiology in the DLEM simulates major physiological processes, such as photosynthesis, autotrophic respiration, allocation among various plant parts (root, stem and leaf), turnover of living biomass, nitrogen uptake and fixation, transpiration, phenology, etc. The soil biogeochemistry component simulates N mineralization, nitrification/denitrification, NH<sub>3</sub> volatilization, leaching of soil mineral N, decomposition and fermentation. Thus the DLEM is able to simultaneously estimate emissions of multiple trace gases (CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O) from soils. The dynamic vegetation component in the DLEM simulates two kinds of processes: the biogeographical redistribution when climate changes, and plant competition and succession during the recovery of vegetation after disturbances. Like most dynamic global vegetation models (DGVMs), the DLEM builds on the concept of plant function type (PFT) to describe vegetation distributions. The DLEM also emphasizes the simulation of managed ecosystems, including agricultural systems, plantation forests and pastures (Ren *et al.*, 2007a,b; Liu *et al.*, 2008; Tian *et al.*, 2010a,c). The DLEM has been used to simu-

late the effects of climate variability and change, atmospheric CO<sub>2</sub>, tropospheric O<sub>3</sub>, land-use change, nitrogen deposition and disturbances (e.g. fire, harvest, hurricanes) on terrestrial water and carbon pools and fluxes in China and the USA, and has been calibrated against field data from various ecosystems including forests, grasslands and croplands. Simulated results with the DLEM have also been evaluated against independent field data (Chen *et al.*, 2006; Ren *et al.*, 2007a,b; Liu *et al.*, 2008; Tian *et al.*, 2010a,c).

In DLEM, the carbon balance of vegetation is determined by the photosynthesis, autotrophic respiration, litterfall (related to tissue turnover rate and leaf phenology) and plant mortality rates. Plants assimilate carbon by photosynthesis, and use this carbon to compensate for carbon loss through maintenance respiration, tissue turnover and reproduction. The photosynthesis module of the DLEM estimates the net C assimilation rate, leaf daytime maintenance respiration rate and gross primary productivity (GPP, g C m<sup>-2</sup> day<sup>-1</sup>). The DLEM model represents a one-layered, two-big leaf model, in which the canopy is composed of sun and shaded fractions and the canopy transmission and absorption of shortwave radiation is based on Beer's law. The photosynthesis rate is first calculated at the leaf level, and then scaled up to canopy level by multiplying by leaf area index (LAI). Photosynthesis is the first process by which most carbon and chemical energy enter ecosystems so it has

critical impacts on ecosystem production. The GPP calculation can be expressed as:

$$\text{GPP}_i = (A_i + R_{d_i}) \times \text{LAI}_i \times \text{dayl} \quad (1)$$

and

$$A_i = f(\text{PPFD}_{\text{leaf}i}, g_i, \text{leafN}_i, T_{\text{day}}, C_a, \text{dayl}), \quad (2)$$

where GPP ( $\text{g C m}^{-2} \text{ day}^{-1}$ ) is the gross ecosystem primary productivity for leaf type  $i$  (sunlit leaf or shaded leaf),  $A$  ( $\text{g s}^{-1} \text{ m}^{-2} \text{ leaf}^{-1}$ ) and  $R_d$  ( $\text{g s}^{-1} \text{ m}^{-2} \text{ leaf}^{-1}$ ) are daytime photosynthesis rate and leaf respiration rate, respectively, LAI is leaf area index, dayl (s) is the length of day time, PPFD ( $\mu\text{mol m}^{-2} \text{ s}^{-1}$ ) is the photosynthetic photon flux density,  $g$  ( $\text{m s}^{-1}$ ) is the stomatal conductance of the leaf to  $\text{CO}_2$  flux,  $T_{\text{day}}$  ( $^{\circ}\text{C}$ ) is daytime temperature,  $C_a$  (p.p.m.v.) is the atmospheric  $\text{CO}_2$  concentration and, leafN ( $\text{g N m}^{-2} \text{ leaf}$ ) is the leaf N content. On the basis of the 'strong optimality' hypothesis (Dewar, 1996), DLEM allocates the leaf N to sunlit fraction and shaded fraction each day according to the relative PPFD absorbed by each fraction, to maximize the rate of photosynthesis. In this study, NPP in an ecosystem and annual net carbon exchange (NCE) of the terrestrial ecosystem with the atmosphere were computed with the following equations:

$$\text{NPP} = \text{GPP} - R_d \quad (3)$$

$$\text{NCE} = \text{NPP} - R_H - E_{\text{NAD}} - E_{\text{AD}} - E_P \quad (4)$$

where  $R_d$  is the plant respiration,  $R_H$  is soil respiration,  $E_{\text{NAD}}$  is the magnitude of carbon loss from a natural disturbance and is assigned as 0 here because of the difficulty of simulation under present conditions,  $E_{\text{AD}}$  is carbon loss during the conversion of natural ecosystems to agricultural land and  $E_P$  is the sum of carbon emission from the decomposition of products. For natural ecosystems,  $E_P$  and  $E_{\text{AD}}$  are equal to 0, and so NCE equals net ecosystem production (NEP). In this study, annual change in carbon storage in forest ecosystems is determined by the accumulation of annual NCE (a negative value means carbon release is greater than carbon uptake which leads to decreased carbon storage, and conversely, a positive value means carbon uptake is greater than carbon release which leads to increased carbon storage). In addition, the agricultural module of the DLEM is not based on natural vegetation but is parameterized against several intensively studied agricultural sites in China (<http://www.cern.ac.cn:8080/>), allowing it to address diverse types of land management (fertilizer, irrigation, rotation, harvest, etc.).

To simulate the detrimental effects of air pollution on ecosystem productivity, an  $\text{O}_3$  module was developed based on previous work (Ollinger *et al.*, 1997; Felzer *et al.*, 2004, 2005), in which the direct effect of  $\text{O}_3$  on photosynthesis and the indirect effect of changing inter-cellular  $\text{CO}_2$  concentration on stomatal conductance were simulated. The ratio of  $\text{O}_3$  injury to photosynthesis is defined as  $\text{O}_{3\text{eff}}$ , similar to Ollinger *et al.* (1997), and the sensitivity coefficient  $\alpha$  (an empirically derived ozone response coefficient) for each different plant functional type is based on the work of Felzer *et al.* (2004). The range of  $\alpha$  is  $2.6 \times 10^{-6} \pm 2.3 \times 10^{-7}$  for hardwoods (based on the value used by

Ollinger *et al.* (1997),  $0.7 \times 10^{-6} \pm 2.45 \times 10^{-7}$  for conifers (based on pines) and  $3.9 \times 10^{-6} \pm 5.27 \times 10^{-7}$  for crops, which was calculated from the empirical model of Reich (1987). Errors are based on the standard deviation of the slope from the dose-response curves and the standard error of mean stomatal conductance:

$$\text{GPP}_{\text{O}_3} = \text{GPP} \times \text{O}_{3\text{eff}} \quad (5)$$

$$\text{O}_{3\text{eff}} = 1 - (\alpha g_s \times \text{AOT}_{40}) \quad (6)$$

$$g_s = f(\text{GPP}_{\text{O}_3}). \quad (7)$$

$\text{GPP}_{\text{O}_3}$  is limited GPP due to the  $\text{O}_3$  effect;  $g_s$  is the stomatal conductance ( $\text{mm s}^{-1}$ );  $\text{AOT}_{40}$  is a cumulative  $\text{O}_3$  index (the accumulated hourly  $\text{O}_3$  dose over a threshold of 40 p.p.b. in p.p.b.  $\text{h}^{-1}$ ), and in this study we use a monthly cumulative index as in the work by Felzer *et al.* (2004). The  $\text{AOT}_{40}$  index has often been used to represent vegetation damage due to  $\text{O}_3$  (Fuhrer *et al.*, 1997). Because of limited data regarding  $\text{O}_3$  concentrations throughout China, we used the modelled  $\text{AOT}_{40}$  values developed by Felzer *et al.* (2005). Yet, our photosynthesis module, based on Farquhar *et al.*'s (1980) model, has the potential ability to incorporate  $\text{O}_3$  concentrations as input similar to Martin *et al.* (2001) and Sitch *et al.* (2007) if  $\text{O}_3$  flux data are available.

## Input data

Input data sets include the following: (1) elevation, slope and aspect maps derived from a 1-km resolution digital elevation data set (<http://eng.wdc.cn:8080/Metadata/index.jsp>); (2) soil data sets (pH, bulk density, depth to bedrock, soil texture represented as the percentage content of loam, sand and silt) derived from the soil map at the scale 1:1 million based on the second national soil survey of China (Tian *et al.*, 2010c); (3) a vegetation map (or land-cover map) from the 2000 land-use map of China (LUCC\_2000) developed from Landsat Enhanced Thematic Mapper (ETM) imagery (Liu *et al.*, 2005); (4) a potential vegetation map, constructed by replacing the croplands of LUCC 2000 with potential vegetation in global potential vegetation maps developed by Ramankutty & Foley (1998); (5) the standard IPCC (Intergovernmental Panel on Climate Change, 2007) historical  $\text{CO}_2$  concentration data set; (6) the annual nitrogen deposition data set (Lu & Tian, 2007); (7) long-term land-use history (cropland and urban distribution of China from 1661 to 2000) (Liu & Tian, 2010); (8) ozone  $\text{AOT}_{40}$  data set (see below for detailed information), and (9) daily climate data (maximum, minimum and average temperature, precipitation and relative humidity). A total of 746 climate stations in China plus 29 stations from surrounding countries were used to produce daily climate data for the time period from 1961 to 2005, using an interpolation method similar to that used by Thornton *et al.* (1997). To account for cropland management, we also used the National Agriculture Database (Statistics Bureau of China from 1978 to 2005), which recorded annual

irrigation areas and fertilizer amounts in each province. All data sets have a spatial resolution of 10 km × 10 km; Climate and AOT<sub>40</sub> data sets were developed on a daily time step, and CO<sub>2</sub> and land-use data sets on a yearly time step.

In this study, the AOT<sub>40</sub> data set was derived from the global historical AOT<sub>40</sub> data sets constructed by Felzer *et al.* (2005). Due to the limited ground O<sub>3</sub> monitoring sites in China and the diverse methods used for monitoring O<sub>3</sub>, it is difficult to develop a historical AOT<sub>40</sub> spatial data set based on the interpolation of site-level data like Felzer *et al.* (2004) did for the USA. However, we used limited field O<sub>3</sub> data sets to validate the simulated AOT<sub>40</sub> (e.g. Wang *et al.*, 2005). The AOT<sub>40</sub> (Fig. 2b) data set shows a significant increase in O<sub>3</sub> concentrations in the 1990s across China's forest ecosystems with the highest rate occurring in mid-north China (MN). Precipitation decreased in most areas of the MN region and in the central-eastern section of north China (Fig. 2c).

### Simulation experiment and implementation

In our study, we designed nine simulation experiments to analyse the effects of O<sub>3</sub> only, climate only and the combined effects of O<sub>3</sub> and climate on NPP and NCE in the forest ecosystems of China; we also assessed the relative contributions of other environmental factors influencing the effects of O<sub>3</sub> and climate (Table 1). Experiment I examined the sole extent of O<sub>3</sub> impacts while other environmental factors were constant (I: O<sub>3</sub>). In experiments II and III, we analysed the contribution of climate variability only (II: Climate) and the combined effect of climate and O<sub>3</sub> variability (III: Clm\_O<sub>3</sub>), respectively. Experiment IV was designed to examine all combined effects representing the real world including O<sub>3</sub> pollution, climate change, fertilization effects of nitrogen deposition and CO<sub>2</sub>, and land-cover/land-use change (LCLUC) (IV: All\_Com). The last five experiments (V to IX) were conducted to assess the relative contribution of each environmental factor on forest NPP and NCE: all combined effects without O<sub>3</sub> (V: No\_O<sub>3</sub>), without LCLUC (VI: No\_LCLUC), without climate (VII: No\_Clm), without nitrogen deposition – Ndep (VIII: No\_Ndep) and without CO<sub>2</sub> (IX: No\_CO<sub>2</sub>).

The model simulation began with an equilibrium run to develop the baseline C, N and water pools for each grid. Then a spin-up of about 100 years was applied if climate variability was included in the simulation experiment. Finally, the model ran in transient mode driven by transient data of climate, O<sub>3</sub>, CO<sub>2</sub>, nitrogen deposition and LCLUC.

## RESULTS

### Temporal variability in annual NPP and NCE

Negative effects of tropospheric O<sub>3</sub> on total NPP and NCE during the study period from 1961–2005 were observed in the simulation experiments (Table 2). We found that elevated O<sub>3</sub> only could lead to 0.7% and 116.3% decreases in annual NPP and annual NCE, respectively, between the 1960s and the 1990s. When considering other environmental factors of climate

change, LCLUC, nitrogen deposition and CO<sub>2</sub> together with O<sub>3</sub> pollution (All\_Com), the total forest NPP gradually increased (15.9%) from 1.44 Tg C year<sup>-1</sup> in the 1960s to 1.67 Tg C year<sup>-1</sup> in a recent 6-year period (2000–05). Without O<sub>3</sub> pollution, however, the total forest NPP estimate ranged from 1.44 to 1.70 Tg C year<sup>-1</sup> (increase 17.5%). Annual variability of NPP under different simulation experiments indicated that O<sub>3</sub> pollution has consistent negative effects on forest production (Fig. 3a); total NPP was reduced by 0.2% to 1.6% from the 1960s to 2000–05.

With O<sub>3</sub> effects only, carbon release from forest ecosystems to the atmosphere continuously increased from 0.004 Pg C year<sup>-1</sup> in the 1960s to 0.018 Pg C year<sup>-1</sup> between 2000–05 (Table 2). China's forest ecosystems were estimated as carbon sinks and sequestered on average about 0.101 Pg C year<sup>-1</sup> over the past 45 years, considering the combined effects of the major environmental factors (All\_Com); however, without O<sub>3</sub> pollution, carbon uptake rates were increased by 3.5% in the 1960s and 12.6% in the 6 years 2000–05.

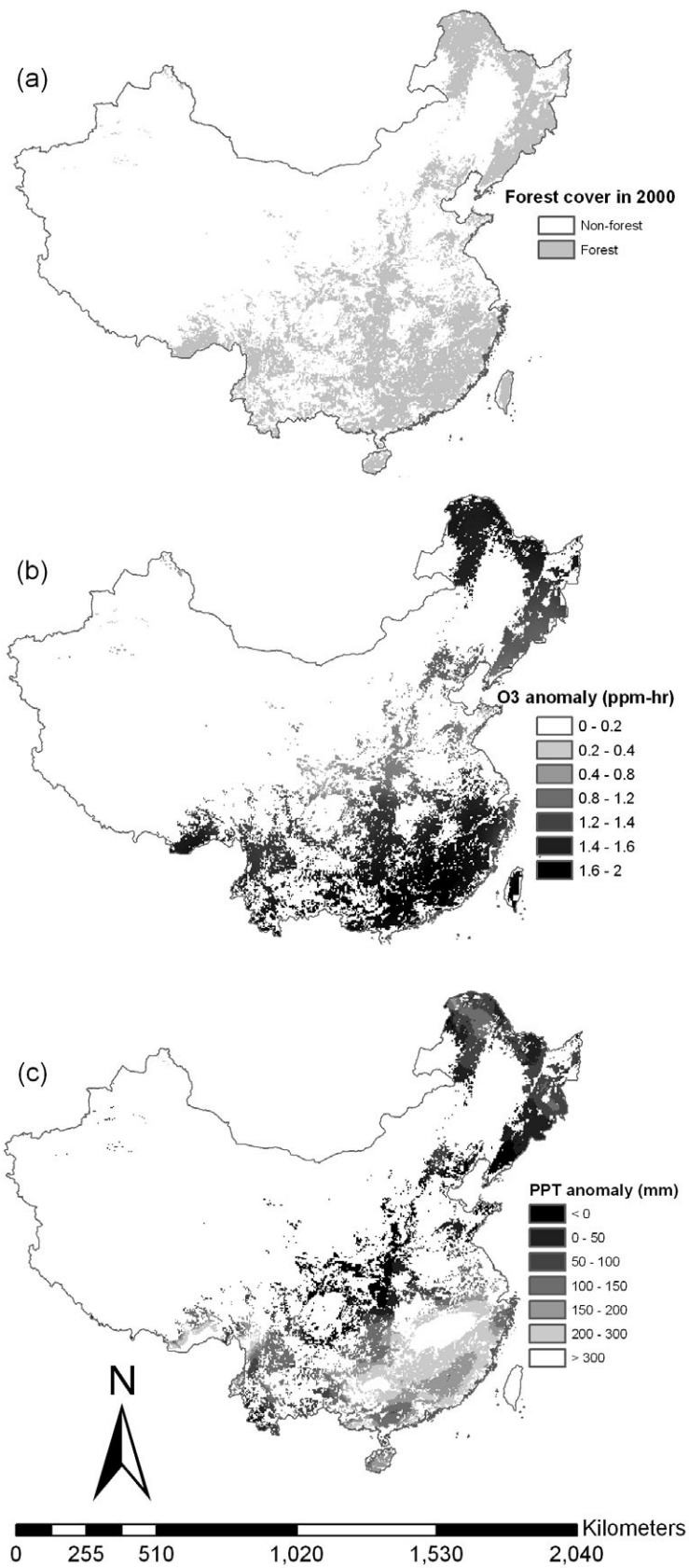
Figure 3 indicates that climate is the dominant factor controlling inter-annual variability in total NPP (Fig. 3a). Climate change/variability could cause carbon uptake or release, and result in inter-annual alterations in NCE (Fig. 3b). Further analysis indicated that the decrease in inter-annual variability of NPP and NCE between 1978 and 1990 was consistent with small inter-annual variability in annual climate (especially in average temperature) and forest area. The offset of the curves in Fig. 3 is largely caused by the combined effects of other major environmental factors including atmospheric CO<sub>2</sub>, nitrogen deposition and LCLUC.

### Spatial variability in annual NPP and NCE

As forests are mostly located in the south-east (SE), south-west (SW) and north-east (NE) regions of China, monsoon climate in most areas and high O<sub>3</sub> in parts of those regions resulted in large spatial variability in NPP and NCE of forest ecosystems (Figs 4 & 5). The highest NPP was distributed in SE China with more than 1500 g C m<sup>-2</sup> year<sup>-1</sup> in some areas of this region. Annual NPP increased across China's forest ecosystems in the 1990s compared to that in the 1960s and the largest increase occurred in SE forest area and then in the NE region (Fig. 4a,b). However, simulated annual NPP was very low (less than 200 g C m<sup>-2</sup> year<sup>-1</sup>) in some parts of the MN region where frequent drought and high O<sub>3</sub> concentrations were experienced. Across China's forest ecosystems, the SE had the largest NCE with a carbon uptake of more than 2000 g C m<sup>-2</sup> between 1961 and 2005, followed by the NE; carbon release appeared in some regions of NE and SW China (Fig. 5).

### Variability of annual NPP and NCE in different forest types

Annual mean NPP and NCE of different PFTs derived from the three simulation experiments indicated that forest types responded differently to increasing O<sub>3</sub> concentration and its



**Figure 2** Map of forest distribution in China (a) and maps of anomalies in the 1990s (relative to the average for 1961–90) for (b) annual average AOT<sub>40</sub> (p.p.b. h<sup>-1</sup>) and (c) precipitation (mm). (Note: AOT<sub>40</sub> is a cumulative O<sub>3</sub> index, the accumulated hourly O<sub>3</sub> dose over a threshold of 40 p.p.b. in p.p.b. h<sup>-1</sup>).

**Table 1** Experimental arrangement.

Simulation experiments		Environmental factors				
		O <sub>3</sub>	Climate	CO <sub>2</sub>	Ndep	LCLUC
I	Only O <sub>3</sub> (O <sub>3</sub> )	H	C	C	C	C
II	Only Climate (Climate)	0	H	C	C	C
III	O <sub>3</sub> _Climate (C <sub>lm</sub> _O <sub>3</sub> )	H	H	C	C	C
IV	O <sub>3</sub> _Climate_LULUC_Ndep_CO <sub>2</sub> (All_Com)	H	H	H	H	H
V	Climate_LULUC_Ndep_CO <sub>2</sub> (No_O <sub>3</sub> )	0	H	H	H	H
VI	O <sub>3</sub> _Climate_Ndep_CO <sub>2</sub> (No_LCLUC)	H	H	H	H	C
VII	O <sub>3</sub> _LCLUC_Ndep_CO <sub>2</sub> (No_C <sub>lm</sub> )	H	C	H	H	H
VIII	O <sub>3</sub> _Climate_LCLUC_CO <sub>2</sub> (No_Ndep)	H	H	H	C	H
IX	O <sub>3</sub> _Climate_LCLUC_Ndep (No_CO <sub>2</sub> )	H	H	C	H	H

Nine simulation experiments include the effect of: only O<sub>3</sub> (I: O<sub>3</sub>); only climate change (II: Climate); combined effects of O<sub>3</sub> and climate (III: C<sub>lm</sub>\_O<sub>3</sub>); all combined effects including O<sub>3</sub>, climate, CO<sub>2</sub> and nitrogen deposition (Ndep) without land-cover/land-use change (LCLUC) (IV: All\_Com); all combined effects without O<sub>3</sub> effects (V: No\_O<sub>3</sub>), without LCLUC (VI: No\_LCLUC), without climate (VII: No\_C<sub>lm</sub>), without Ndep (VIII: No\_Ndep) and without CO<sub>2</sub> (IX: No\_CO<sub>2</sub>). (Note: H is historical data, 0 means no such data and C is constant data. Here we use CO<sub>2</sub> concentration (296 p.p.m.) in 1900 and mean climate data sets in the 30 years from 1961 to 1990 and the potential vegetation map as constant values.)

**Table 2** Decadal mean and change rates of average annual net primary productivity (NPP) and net carbon exchange (NCE) in the 1960s, 1990s and 2000–05 under the combined effects with and without O<sub>3</sub> pollution.

	NPP				NCE			
	O <sub>3</sub> (Pg C year <sup>-1</sup> , 10 <sup>15</sup> g C year <sup>-1</sup> )	All_Com	No_O <sub>3</sub>	All_Com– No_O <sub>3</sub> (%)	O <sub>3</sub> (Pg C year <sup>-1</sup> , 10 <sup>15</sup> g C year <sup>-1</sup> )	All_Com	No_O <sub>3</sub>	All_Com– No_O <sub>3</sub> (%)
1960s	1.30	1.44	1.44	–0.2	–0.004	0.081	0.083	–3.5
1990s	1.29	1.60	1.61	–0.9	–0.008	0.112	0.123	–8.6
2000–05	1.28	1.67	1.70	–1.6	–0.018	0.121	0.138	–12.6
1990s–1960s %	–0.7	11.1	11.9		–116.3	38.6	46.3	
2000–2005–1960s %	–1.2	15.9	17.5		–371.8	49.3	64.8	

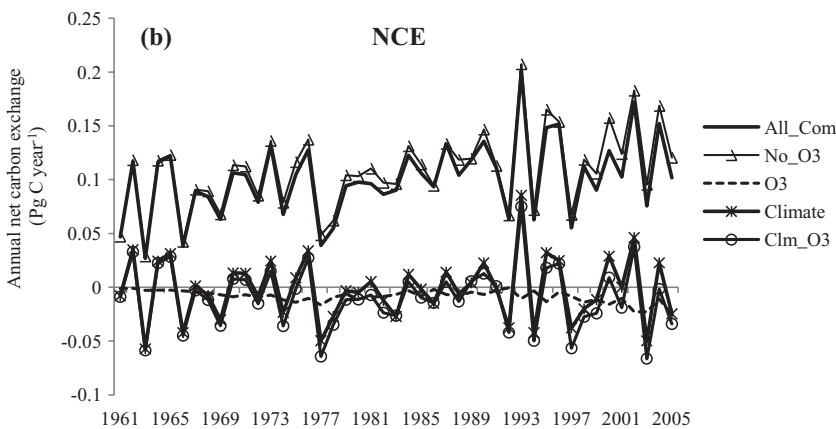
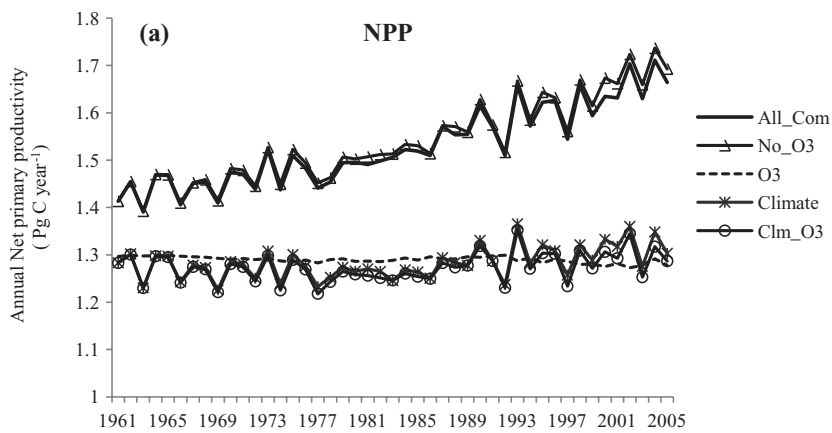
The simulation experiment All\_Com addresses the combined effects of all environmental factors including O<sub>3</sub>, climate, land-cover/land-use change, nitrogen deposition and CO<sub>2</sub>; the simulation experiment No\_O<sub>3</sub> addresses the combined effects of all environmental factors except O<sub>3</sub>.

interaction with other environmental factors (Table 3). From 1961 to 2005, the annual mean forest NPP resulting from all combined effects with O<sub>3</sub> exposure (All\_Com) ranged from 0.003 Pg C year<sup>-1</sup> in the boreal broadleaf deciduous forest to 0.680 Pg C year<sup>-1</sup> in temperate needleleaf evergreen forest; accordingly, the range varied from 0.003 to 0.681 Pg C year<sup>-1</sup> under the simulation experiment of all combined effects without O<sub>3</sub> exposure (No\_O<sub>3</sub>). At the same time, annual carbon sequestration rates (NCE) ranged from 0.1 (based on All\_Com) or 0.2 (based on No\_O<sub>3</sub>) Tg C year<sup>-1</sup> in tropical broadleaf deciduous forest to 53.5 (based on All\_Com) or 53.8 (based on No\_O<sub>3</sub>) Tg C year<sup>-1</sup> in temperate needleleaf evergreen forest. Comparing simulations of O<sub>3</sub> exposure with those without O<sub>3</sub> exposure, both annual NPP and NCE in different forest types decreased from 0.1% (NPP) and 0.4% (NCE) for temperate needleleaf evergreen forest down to 2.6% (NPP) and 43.1% (NCE) for tropical broadleaf deciduous forest. Differing from

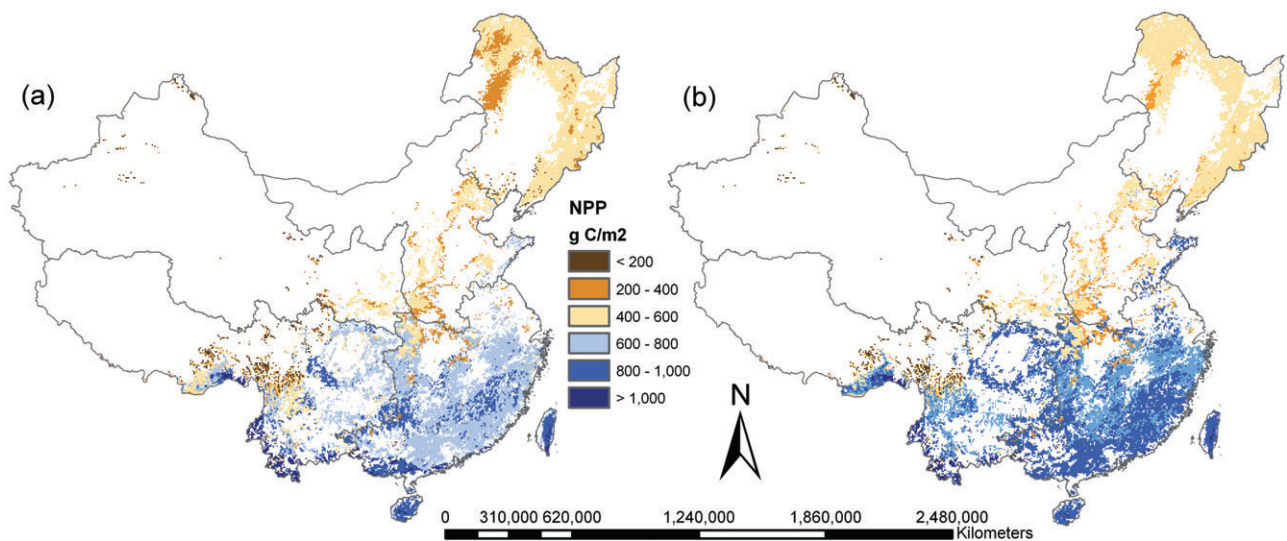
the combined effect with O<sub>3</sub> (All\_Com) and without O<sub>3</sub> (No\_O<sub>3</sub>), the O<sub>3</sub>-only (O<sub>3</sub>) released C in all different forest types with a peak release rate of 6.7 Tg C year<sup>-1</sup>, which means that the temperate broadleaf deciduous forest was more sensitive to O<sub>3</sub> than other types of forests. This could be because most temperate broadleaf deciduous forests are located in the places that experienced high O<sub>3</sub> levels and also because the O<sub>3</sub>-exposure coefficient for this forest type is higher than other forest types, given the model constraints.

#### Relative contributions of O<sub>3</sub> pollution and climate change to NPP and NCE

Temporal variations of relative contributions (Fig. 6) indicated that O<sub>3</sub> pollution had negative effects on both NPP and NCE during the period 1961–2005, with two periods (between the late 1970s and the early 1980s and after the 1990s) of high



**Figure 3** (a) Changes of annual net primary productivity (NPP; Pg C year<sup>-1</sup>) and (b) annual net carbon exchange (NCE; Pg C year<sup>-1</sup>) between forest ecosystems and atmosphere during 1961–2005 under different simulation experiments. Note: the All\_Com scenario considers all the environmental factors including O<sub>3</sub> pollution, climate, nitrogen deposition, CO<sub>2</sub>, land-cover and land-use change; No\_O<sub>3</sub> means the All\_Com without O<sub>3</sub> effects; O<sub>3</sub>, Climate and Clm\_O<sub>3</sub> are simulation experiments of O<sub>3</sub> only, climate only and a combination of climate and O<sub>3</sub>.

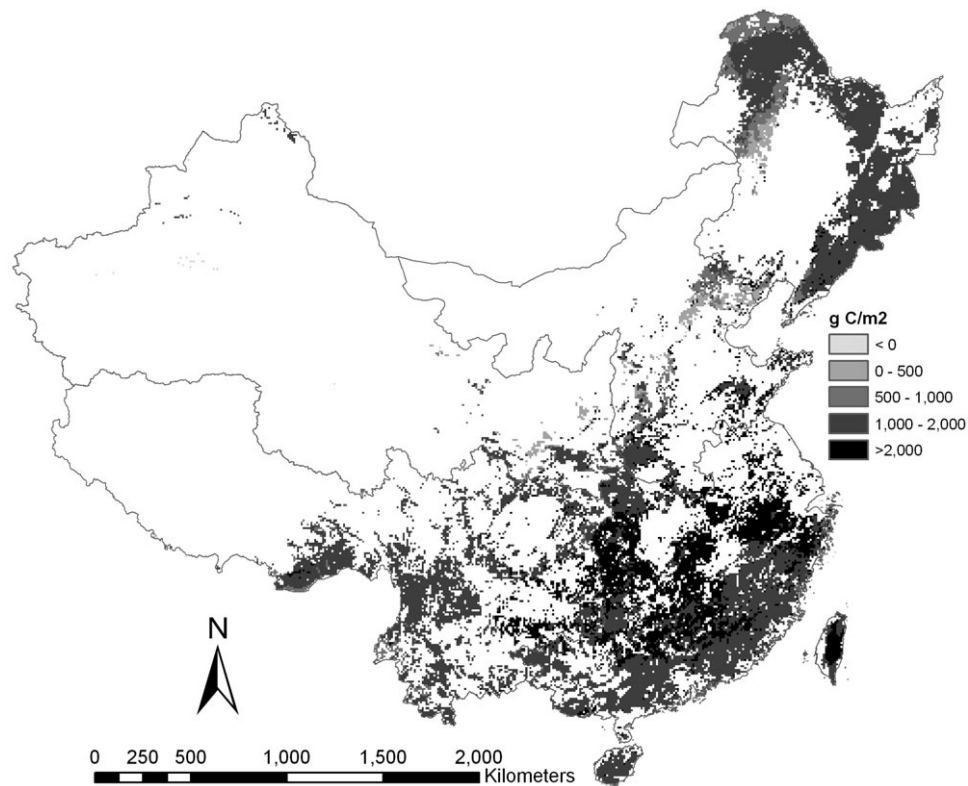


**Figure 4** Decadal mean net primary productivity (NPP) (g C m<sup>-2</sup> year<sup>-1</sup>) in the 1960s (a) and in the 1990s (b) under the All\_Com simulation experiments considering O<sub>3</sub> pollution, climate change, atmospheric CO<sub>2</sub>, nitrogen deposition and land-cover/land-use change.

reductions in NPP and NCE. Climate change had both negative and positive effects on NPP and NCE, and was the major factor controlling the inter-annual variability of NPP and NCE. Between the late 1970s and the early 1980s, climate change had continuously reduced NPP and NCE, while after the 1990s it

contributed greatly to the increase in NPP. The negative effects of O<sub>3</sub> pollution were either accelerated or offset by climate change, therefore the interactive effects of the two showed continuously aggregated effects on the reductions in NPP and NCE between the late 1970s and 1990s.



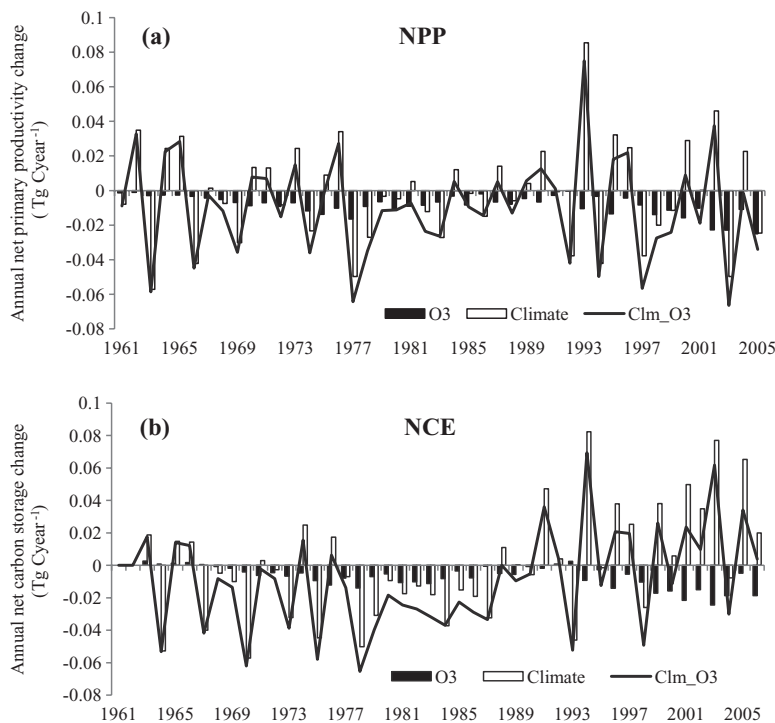


**Figure 5** The total accumulated net carbon exchange (net carbon storage) ( $\text{g C m}^{-2}$ ) in forest ecosystems during the period 1961–2005 under the All\_Com simulation experiment considering O<sub>3</sub> pollution, climate change, atmospheric CO<sub>2</sub>, nitrogen deposition and land-cover/land-use change.

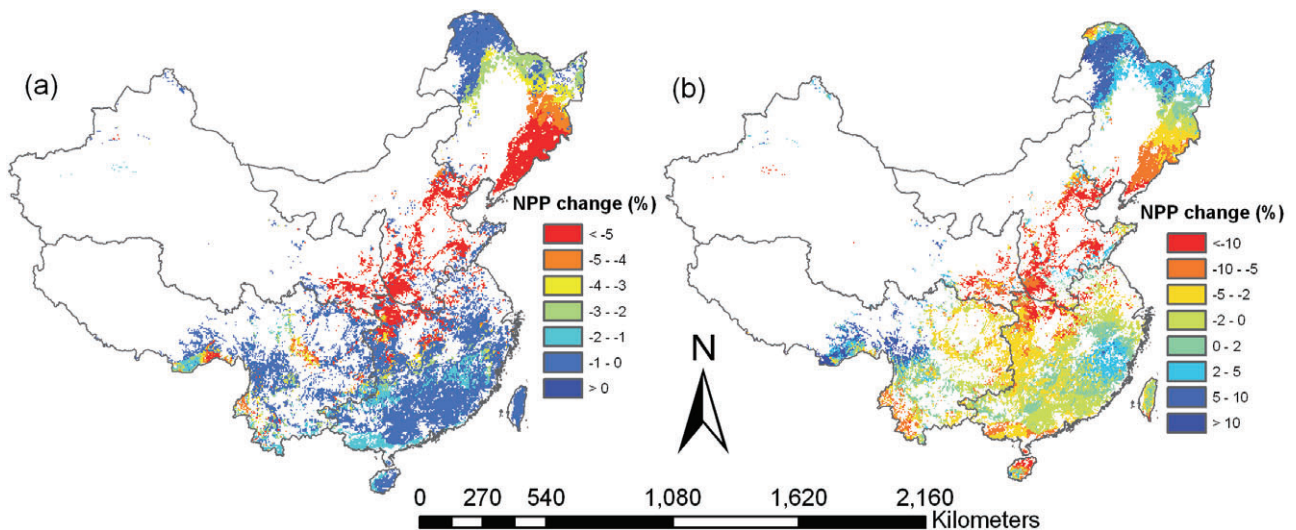
**Table 3** Average annual net primary productivity (NPP) and carbon storage in different forest types during the period 1961–2005 across China's forest ecosystems under the simulation experiments of the combined effects with (All) and without O<sub>3</sub> pollution (No\_O<sub>3</sub>) and effects of O<sub>3</sub> only (O<sub>3</sub>).

NPP	Simulation experiments			
	All_Com	No_O <sub>3</sub>	O <sub>3</sub>	All_Com–No_O <sub>3</sub>
	Pg C year <sup>-1</sup> (= 10 <sup>15</sup> gC year <sup>-1</sup> )			(%)
Boreal broadleaf deciduous forest	0.003	0.003	0.003	–1.1
Boreal needleleaf deciduous forest	0.075	0.075	0.067	–0.3
Temperate broadleaf deciduous forest	0.340	0.349	0.285	–2.3
Temperate broadleaf evergreen forest	0.242	0.244	0.209	–0.6
Temperate needleleaf evergreen forest	0.680	0.681	0.557	–0.1
Temperate needleleaf deciduous forest	0.030	0.031	0.026	–0.4
Tropical broadleaf deciduous forest	0.003	0.004	0.003	–2.6
Tropical broadleaf evergreen forest	0.152	0.153	0.137	–0.6
	Tg C year <sup>-1</sup> (= 10 <sup>12</sup> C year <sup>-1</sup> )			(%)
Boreal broadleaf deciduous forest	0.1	0.2	–0.0	–21.8
Boreal needleleaf deciduous forest	3.5	3.7	–0.2	–5.2
Temperate broadleaf deciduous forest	22.9	29.0	–6.7	–21.1
Temperate broadleaf evergreen forest	13.1	14.1	–0.7	–7.6
Temperate needleleaf evergreen forest	53.5	53.8	–0.2	–0.4
Temperate needleleaf deciduous forest	1.6	1.7	–0.1	–6.2
Tropical broadleaf deciduous forest	0.1	0.1	–0.1	–43.1
Tropical broadleaf evergreen forest	5.7	6.3	–0.1	–9.6
Forest ecosystems	100.6	109.0	–8.5	–7.7

The All\_Com addresses the combined effects of all environmental factors including O<sub>3</sub>, climate, land-cover/land-use change, nitrogen deposition and CO<sub>2</sub>; the No\_O<sub>3</sub> addresses the combined effects of all environmental factors except O<sub>3</sub>.



**Figure 6** Annual changes in relative contributions of ozone pollution, climate change and their interaction to (a) annual net primary productivity (NPP) and (b) net carbon storage (Tg C year<sup>-1</sup>). Note: O<sub>3</sub>, Climate and Clm\_O<sub>3</sub> are simulation experiments of O<sub>3</sub> only, climate only and a combination of climate and O<sub>3</sub>.

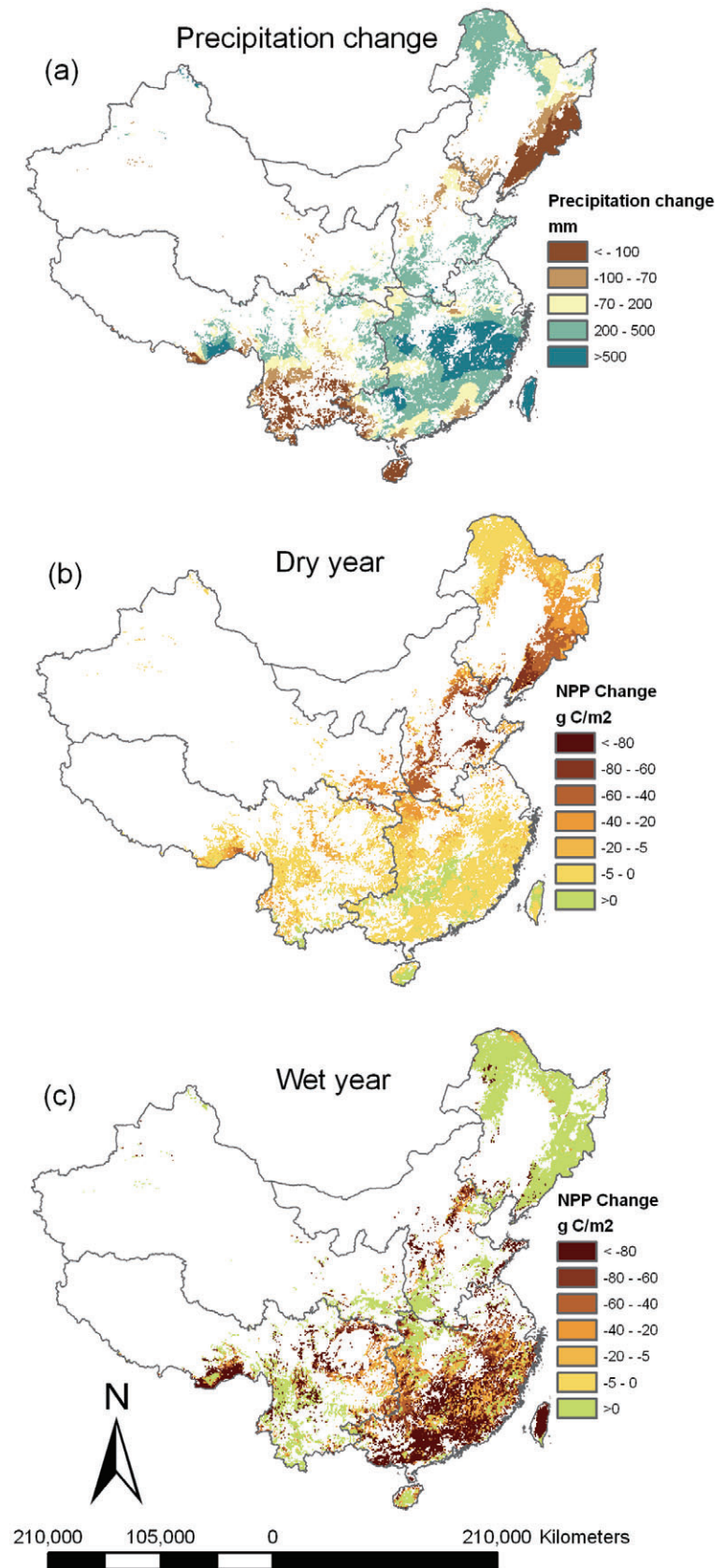


**Figure 7** Rate of change of net primary productivity (NPP; %) between the 1990s and 1960s under scenarios of (a) O<sub>3</sub> only and (b) O<sub>3</sub> + Climate.

Similarly, we found that spatial patterns of NPP that changed under only O<sub>3</sub> pollution effects between the 1990s and 1960s varied from region to region (Fig. 7a), with the rates of reduction ranging from less than 0% to 5%. Combined with climate effects, the rate of decrease was increased or decreased ranging from less than -10% to 10% (Fig. 7b), which indicated that climate change was the dominant factor in controlling changes in spatial patterns. In a further analysis, we found that the spatial patterns were strongly consistent with the precipitation patterns (Fig. 2c). NPP decreased by more than 15% in some areas of the NE and MN regions that were experiencing low precipitation

and high O<sub>3</sub> concentrations, and increased more than 10% in some areas of SE China's forestlands, where high precipitation and low O<sub>3</sub> concentrations were found.

Under extreme climate conditions (wet years and dry years, which were defined simply according to the annual total precipitation in this study), we examined how the O<sub>3</sub>-induced NPP change can be influenced by extreme climate change. Here we investigated the effects of O<sub>3</sub> only combined with the interactive effects of O<sub>3</sub> and climate while removing the dominant effects of climate, in which NPP differences were calculated between NPP derived from O<sub>3</sub> + climate and from climate only in dry (Fig. 8b)



**Figure 8** (a) Annual total precipitation difference between wet and dry years. (b) Annual total NPP change induced by O<sub>3</sub> pollution in dry years and wet years (difference between the O<sub>3</sub> + Climate and the climate-only simulation experiments).

**Table 4** Estimations of mean net primary productivity (NPP), total NPP and carbon sequestration rate in different forest types and the whole forest ecosystems using models, inventory-based and remote sensing (RS)-based methods.

Forests type	Method	Period	Mean NPP Mg ha <sup>-1</sup> year <sup>-1</sup>	Reference
Boreal broadleaf deciduous forest	Biomass	1989–93	5.4–15.1	Ni, 2001
	RS-based estimation	1994	6.3	Jiang <i>et al.</i> (1999)
	Process-based DLEM model	1989–93	4.3	This study
		1994	4.4	
Boreal needleleaf deciduous forest	Biomass	1989–93	5.4–15.1	Ni, 2001
	RS-based estimation	1994	6.7	Jiang <i>et al.</i> (1999)
	Measured	1994	6.5–8.5	This study
	Process-based DLEM model	1989–93	5.5	
		1994	5.6	
Temperate broadleaf deciduous forest	Biomass	1989–93	8.7–23.1	Ni, 2001
	RS-based estimation	1994	9.0–12.4	Jiang <i>et al.</i> (1999)
	Measured	1994	10.5	This study
	Chikugo model	1994	10.7	
	Process-based DLEM model	1989–93	8.2	
		1994	8.3	
Temperate broadleaf evergreen forest	Biomass	1989–93	8.5–13.7	Ni, 2001
	RS-based estimation	1994	10.1–17.4	Jiang <i>et al.</i> (1999)
	Process-based model	–	8.90	Xiao <i>et al.</i> (1998)
	Process-based model	1989–93	8.9	This study
		1994	9.1	
		45-year average	7.2 ± 0.4	

and wet years (Fig. 8c), respectively. Our results indicated that the O<sub>3</sub>-only simulation experiment and its combination with wet conditions could result in a greater reduction in NPP compared with the same simulation experiment with dry conditions, with the greatest reduction (> 80 g C m<sup>-2</sup>) occurring in some areas of the southern and eastern regions. However, increases in NPP occurred in some parts of the SE and SW regions. The pattern of NPP change was very consistent with the distribution of precipitation change (the precipitation difference between wet years and dry years) (Fig. 8a).

## DISCUSSION

### Estimates of NPP and NCE

Our estimations of annual mean NPP were similar to those of other studies from inventory-based (Ni, 2001) and remote sensing (RS)-based estimations (Jiang *et al.*, 1999) and other process-based models (Xiao *et al.*, 1998) (Table 4). For example, the simulated annual NPP of 5.5 Mg ha<sup>-1</sup> year<sup>-1</sup> in boreal needleleaf deciduous forest and 8.9 Mg ha<sup>-1</sup> year<sup>-1</sup> in temperate broadleaf evergreen forests were comparable to 5.4–15.1 Mg ha<sup>-1</sup> year<sup>-1</sup> and 8.5–17.4 Mg ha<sup>-1</sup> year<sup>-1</sup> for the two forest types from biomass inventory-based estimations (Ni, 2001). The total carbon sequestration rate (or NCE) of 0.126–0.137 Pg C year<sup>-1</sup> during 1981–2000 was very similar to the estimation by Fang *et al.* (2007), which indicates that China's forests were a carbon sink as assessed by both inventory data and

process-based models (Table 5). However, the estimations of total annual NPP varied among different studies (e.g. Running *et al.*, 2004; Zhuang *et al.*, 2009) ranging from 0.778 to 1.514 Pg C year<sup>-1</sup>. The uncertainty in NPP estimates may be caused by differences in research methods and forest area estimates. For example, methods based on forest inventory or statistical/empirical models depend on plot numbers, and whether their spatial distribution is homogeneous or not; consequently, there are different estimates derived from different inventory data, regression equations and influencing indicators.

### Estimates of ozone impacts

Our results, which indicate that O<sub>3</sub> had negative effects on forest ecosystem production and carbon storage, are similar to short- and long-term studies on the effects of O<sub>3</sub> based on field experiments (e.g. Saleem *et al.*, 2001; Chappelka, 2002) and model simulations (e.g. Ollinger *et al.*, 1997, 2002; Martin *et al.*, 2001; Felzer *et al.*, 2004; Hanson *et al.*, 2005) in the USA and Europe. Direct ambient O<sub>3</sub> damage could reduce forest productivity by 1–10% in both Europe (Broadmeadow, 1998) and the USA (Chappelka & Samuelson, 1998), and the total biomass of trees could be significantly reduced by 7% due to current ambient O<sub>3</sub> (40 p.p.b. on average), compared with trees in charcoal-filtered (pristine environments) controls across Northern Hemisphere forests (Wittig *et al.*, 2009). Our estimates were similar to previous work ranging from 0 to an 11.8% reduction.

**Table 5** Estimations of total net primary productivity (NPP) and net carbon exchange (NCE) of forest ecosystems at national level using models, inventory-based, geospatial statistical and remote sensing (RS)-based methods.

National	Method	Period	Area (10 <sup>6</sup> square kilometres)	NPP (Pg Cyr <sup>-1</sup> ) NCE (Pg Cyr <sup>-1</sup> )	Reference
NPP	RS-based	1981–99	–	1.035	Running <i>et al.</i> (2004)
	Geospatial, regression	1970–94	1.57	1.325 ± 0.102	Zhuang <i>et al.</i> (2009)
	Statistical, kriging	1970–94	1.57	1.258 ± 0.186	Zhuang <i>et al.</i> (2009)
	Process-based model	1901–2002	1.57	0.778	Zhuang <i>et al.</i> (2009)
	Process-based model	1970–94	1.26–1.37	1.514 ± 0.054	This study
NCE	Inventory data	1981–2000	1.17–1.43	0.115–0.145	Fang <i>et al.</i> (2007)
	Process-based model	1981–2000	1.26–1.37	0.126–0.137	This study

### Interactive effects of ozone pollution and climate change

Our estimates of O<sub>3</sub>-induced reduction in NPP and NCE include the direct contribution of elevated tropospheric O<sub>3</sub> damage and its interactive effects with other environmental factors (Fig. 3a,b). Our sensitivity experiments indicated that climate variability could offset or exacerbate the O<sub>3</sub>-induced reduction in forest productivity and carbon storage, and also lead to substantial inter-annual variability in annual NPP and annual NCE (Fig. 6a,b). Between the 1960s and the 1990s, the combined effects of O<sub>3</sub> pollution and climate change/variability resulted in a much higher reduction in NPP across China's forest ecosystems than the O<sub>3</sub> pollution only did. Another simulation experiment we conducted indicated that the O<sub>3</sub>-induced reduction in NPP in a wet year was higher than that in a dry year (Fig. 8), which was consistent with previous studies (e.g. Tingey & Hogsett, 1985); however, the reasons for these responses are different. In the study conducted by Tingey & Hogsett (1985), high O<sub>3</sub>-induced reduction in NPP in wet conditions occurred because of greater O<sub>3</sub> uptake with increased stomatal conductance caused by high soil moisture and atmospheric humidity (Sitch *et al.*, 2007; Felzer *et al.*, 2009). As our modelling study here used the O<sub>3</sub> AOT<sub>40</sub> index rather than O<sub>3</sub> fluxes through stomatal conductance, the large reduction in NPP in wet conditions in our study was due to a relatively higher NPP in wet conditions than in dry conditions, which is similar to Felzer *et al.*'s (2005) estimates regarding significantly higher O<sub>3</sub>-induced crop yield reductions when considering fertilizer application than without considering fertilizer application. A few experiments in mature forests indicated that an elevated O<sub>3</sub> level could decrease transpiration, reduce photosynthesis and thus decrease NPP (Wittig *et al.*, 2007). However, McLaughlin *et al.* (2007) hypothesized that the impaired stomata due to elevated O<sub>3</sub> exposure could lead to increased transpiration, which suggested that it is important to explore the underlying mechanisms regarding the effect of O<sub>3</sub> on ecological processes linking water and carbon cycles between ecosystems and the atmosphere (Sitch *et al.*, 2007). Therefore, more field experiments and observations are needed to address O<sub>3</sub> fluxes and their direct influence on physiological and ecosystem processes such

as stomatal conductance, photosynthesis and carbon allocation for modelling studies at a regional level.

### Fertilization effects of CO<sub>2</sub> and nitrogen deposition

We found that the continuous increases in NPP and NCE after the late 1980s were mainly caused by the fertilization effects of increasing CO<sub>2</sub> and nitrogen deposition (Fig. 3a,b). In order to examine how CO<sub>2</sub> and nitrogen fertilization offset the O<sub>3</sub> damage along with climate change or LCLUC, we conducted other four simulation experiments (VI to IX). Together with experiment V, we estimated the relative contributions of individual O<sub>3</sub>, climate, CO<sub>2</sub>, nitrogen deposition and LCLUC. Without considering LCLUC, we found that an O<sub>3</sub>-climate-induced carbon source could be converted to a carbon sink by including the effects of CO<sub>2</sub> and nitrogen deposition. An increase of about 48% in NPP occurred in the CO<sub>2</sub>-N simulation experiment, which was consistent with previous studies (e.g. Ollinger *et al.*, 2002; Felzer *et al.*, 2004; Hanson *et al.*, 2005). Considering LCLUC in the model, we found that CO<sub>2</sub> and nitrogen deposition contributed to an increase in NPP and carbon sequestration during the period 1961–2005, which could offset the combined negative effects of O<sub>3</sub> pollution, climate change and LCLUC on annual NPP. These results were similar to those for NCE; either CO<sub>2</sub> or nitrogen deposition could offset the combined negative effects of O<sub>3</sub> pollution and LCLUC on annual NCE using a 45-year (1961–2005) average (Table 6). Compared to Ollinger *et al.*'s (2002) work in which there was no climate change, and the study by Felzer *et al.* (2004) which did not consider nitrogen deposition, our simulations include all major environmental factors including O<sub>3</sub>, climate, nitrogen deposition, CO<sub>2</sub>, and LCLUC. We found that nitrogen deposition alone could compensate for the combined negative effects of O<sub>3</sub> and LCLUC in China, while nitrogen deposition can only offset the negative effects of O<sub>3</sub> exposure in the north-eastern USA (Ollinger *et al.*, 2002), which indicated that nitrogen deposition played an important role in enhancing carbon uptake by China's forest ecosystems. However, we further found that excessive nitrogen or nitrogen depletion caused by CO<sub>2</sub> enrichment did not change the decrease in forest production induced by O<sub>3</sub> and climate; instead, the nitrogen and water use efficien-

**Table 6** Response of forest net primary productivity (NPP) and net carbon exchange (NCE) to ambient O<sub>3</sub>, O<sub>3</sub> combined with climate, O<sub>3</sub> combined with climate, nitrogen deposition (Ndep) and CO<sub>2</sub> with consideration of land-cover/land-use change (LCLUC) using different models and inventory-based methods.

	Period	Location	NPP / NCE change rate (%)	Reference
Ambient O <sub>3</sub>	–	USA and Europe	NPP: –1 to –10%	Broadmeadow (1998), Chappelka & Samuelson (1998)
	–	Northern Hemisphere	Biomass: –7%	Wittig <i>et al.</i> (2009)
	1961–2005	China	NPP: 0 to –11.8%	This study
O <sub>3</sub> + climate	1987–92	North-eastern USA	NPP: –3% to –16% or (mean –7.4%) NPP: –3.8%	Ollinger <i>et al.</i> (1997) Felzer <i>et al.</i> (2004)
		China	NPP: –2% to –9%	This study
O <sub>3</sub> + Climate + LCLUC + CO <sub>2</sub> + Ndep	1860–1995	North-eastern USA	NCE: –O <sub>3</sub> ≈ +Ndep	Ollinger <i>et al.</i> (2002)
			NPP: –(O <sub>3</sub> , LCLUC <sup>1</sup> ) ≈ +(Ndep, CO <sub>2</sub> )	
	1961–2005	China	NPP: –(O <sub>3</sub> , climate, LCLUC <sup>2</sup> ) ≈ +(CO <sub>2</sub> ) NPP: –(O <sub>3</sub> , climate, LCLUC <sup>3</sup> ) ≈ +(Ndep, CO <sub>2</sub> ) NCE: –(O <sub>3</sub> , LCLUC <sup>1</sup> ) ≈ +(CO <sub>2</sub> ) < +(Ndep)	Felzer <i>et al.</i> (2004) This study

LCLUC<sup>1</sup> includes land-cover change with timber harvest, agriculture and management; LCLUC<sup>2</sup> means land-cover change without agricultural management and timber harvest; LCLUC<sup>3</sup> includes land-cover change with agricultural management; Ndep is nitrogen deposition.

cies were reduced (Tian *et al.*, 2010a,c). In addition, the reduction of excessive nitrogen deposition could also reduce O<sub>3</sub> concentrations due to the fact that O<sub>3</sub> is produced through the photochemical cycle via various reactions with N and volatile organic compounds (VOCs). In order to address the complex interactions among multiple environmental stresses such as O<sub>3</sub>, climate, CO<sub>2</sub>, nitrogen deposition and LCLUC at regional scales, we need to conduct multifactor field experiments and establish long-term observational network for monitoring various atmospheric compositions (CO<sub>2</sub>, O<sub>3</sub>, N deposition) and LCLUC.

### Future research needs

This study provides the first estimate of how elevated tropospheric O<sub>3</sub> and climate change/variability have affected forest productivity and carbon sequestration in China. To accurately assess the impacts of O<sub>3</sub> pollution and climate change on forest ecosystems, it is of critical importance to improve the existing input data for driving ecosystem models and better address the representation of ecological processes in response to O<sub>3</sub> exposures in the model. First of all, the current major input data (e.g. O<sub>3</sub> and forest age) need to be further improved by incorporating available observational data. For example, the regional database illustrating the increased gradient of the O<sub>3</sub> AOT<sub>40</sub> index from SE to NW China needs to be evaluated using more site observations. Since China's massive afforestation efforts in the past three decades have resulted in a rapid increase in younger forests (Sun *et al.*, 2006), detailed information on age-related forest coverage is needed to better address O<sub>3</sub> and climate effects, because mid-rotation and early growth plantations are more vulnerable to external environmental stressors such as drought and O<sub>3</sub> than mature trees. Additionally, multifactorial field experiments on forests in China are urgently needed and could provide essential information for parameterizing and validating ecosystem models in order to better address the interactive

effects of O<sub>3</sub> with climate and other environmental stresses. The model needs improvement based on available information; for example, this current version of the DLEM model has no explicit function to address the allowance for O<sub>3</sub> damage healing due to cellular repair and the replacement or addition of new leaves without proper parameters against sustainable observations, which would somewhat ameliorate the effects of the impact on estimated forest carbon sequestration. If O<sub>3</sub> flux data and field observations are available, studies similar to the work of Sitch *et al.* (2007) are useful and important to gain a better understanding of the dynamics of ecosystem structure and functioning. In addition, further detailed evaluations of model results in this study related to O<sub>3</sub> changes and the combined effects of O<sub>3</sub> and climate (e.g. daily or monthly patterns in different places) are needed with accessible databases.

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