



## Effects of tropospheric ozone pollution on net primary productivity and carbon storage in terrestrial ecosystems of China

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Received 8 February 2007; revised 8 May 2007; accepted 15 October 2007; published 17 November 2007.

[1] We investigated the potential effects of elevated ozone (O<sub>3</sub>) along with climate variability, increasing CO<sub>2</sub>, and land use change on net primary productivity (NPP) and carbon storage in China's terrestrial ecosystems for the period 1961–2000 with a process-based Dynamic Land Ecosystem Model (DLEM) forced by the gridded data of historical tropospheric O<sub>3</sub> and other environmental factors. The simulated results showed that elevated O<sub>3</sub> could result in a mean 4.5% reduction in NPP and 0.9% reduction in total carbon storage nationwide from 1961 to 2000. The reduction of carbon storage varied from 0.1 Tg C to 312 Tg C (a decreased rate ranging from 0.2% to 6.9%) among plant functional types. The effects of tropospheric O<sub>3</sub> on NPP were strongest in east-central China. Significant reductions in NPP occurred in northeastern and central China where a large proportion of cropland is distributed. The O<sub>3</sub> effects on carbon fluxes and storage are dependent upon other environmental factors. Therefore direct and indirect effects of O<sub>3</sub>, as well as interactive effects with other environmental factors, should be taken into account in order to accurately assess the regional carbon budget in China. The results showed that the adverse influences of increasing O<sub>3</sub> concentration across China on NPP could be an important disturbance factor on carbon storage in the near future, and the improvement of air quality in China could enhance the capability of China's terrestrial ecosystems to sequester more atmospheric CO<sub>2</sub>. Our estimation of O<sub>3</sub> impacts on NPP and carbon storage in China, however, must be used with caution because of the limitation of historical tropospheric O<sub>3</sub> data and other uncertainties associated with model parameters and field experiments.

**Citation:** 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, *J. Geophys. Res.*, 112, D22S09, doi:10.1029/2007JD008521.

### 1. Introduction

[2] The tropospheric ozone (O<sub>3</sub>) level has been increasing across a range of scales: local, national, continental, and even global [e.g., Akimoto, 2003]. Tropospheric O<sub>3</sub> levels might increase substantially in the future [Streets and Waldhoff, 2000]. Advection from the Asian continent increases pollutant levels over the Pacific Ocean [Jacob et al., 1999; Mauzerall et al., 2000], and eventually influences North America and Europe by intercontinental transport [Jaffe et al., 2003; Wild and Akimoto, 2001]. Ozone can influence both ecosystem structure and functions [e.g., Heagle, 1989; Heagle et al., 1999; Ashmore, 2005; Muntifering et al., 2006]. Over 90% of vegetation damage may be the result of tropospheric ozone alone, and it could

cause reductions in crop yield and forest production ranging from 0% to 30% [Adams et al., 1989]. Approximately 50% of forests might be exposed to higher O<sub>3</sub> level (>60 ppb) by 2100. Therefore there is an urgent need to investigate the adverse effects of O<sub>3</sub> on terrestrial ecosystem production.

[3] Air pollution is one of the most pressing environmental concerns in China [Liu and Diamond, 2005]. The rapid urbanization and industrialization, and intensive agricultural management in the past decades, are closely related to increasing fossil fuel combustion and fertilizer application. Between 1980 and 1995, fertilizer use in China was 36% higher than the average in developed countries (where fertilizer use has been decreasing), and 65% higher than the average in developing countries [Aunan et al., 2000]. Both fossil fuel consumption and N-fertilizer application will highly contribute to total emissions of NO<sub>x</sub>, a main O<sub>3</sub> precursor, and consequently result in increased atmospheric O<sub>3</sub> concentration. It was estimated that China's emissions of NO<sub>x</sub> might increase by a factor of four toward the year 2020, compared to the emissions in 1990 under a non-control scenario [van Aardenne et al., 1999], which would lead to a much larger increase of surface O<sub>3</sub> with 150 ppb

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level of ozone in some locations [Elliott *et al.*, 1997]. Consequently, it is important to study the impacts of O<sub>3</sub> on terrestrial ecosystems in China. Although studies on ozone have been carried out in China for about 20 a, observations of O<sub>3</sub> concentrations are still limited, and the records of most sites are discontinuous [e.g., Chameides *et al.*, 1999; Liu *et al.*, 2004; Wang *et al.*, 2007]. Several experiments demonstrated the interaction of O<sub>3</sub> and CO<sub>2</sub> on locally grown species and cultivars in China [e.g., Wang, 1995; Wang *et al.*, 2002; Guo *et al.*, 2001; Bai *et al.*, 2003]. However, these studies rarely involved other plant functional types (PFTs), such as forests and grassland. An assessment of O<sub>3</sub> effects on different PFTs at a large scale over a long time period has not been done yet. To illustrate the O<sub>3</sub> effects at a continental scale, it is necessary to consider interactive effects of O<sub>3</sub> with other environmental factors on terrestrial ecosystem production and carbon storage.

[4] Quantitative assessment of O<sub>3</sub> effects on terrestrial ecosystem production has been conducted since the 1980s on the basis of empirical or process-based dynamic simulations [Ren and Tian, 2007]. Well-documented empirical models, such as the Weibull function, are based on exposure indices and corresponding exposure-response relationships, and have been used to assess crop and forest production loss, as well as economic losses [e.g., Heck *et al.*, 1984; Heggstad and Lesser, 1990; Chameides *et al.*, 1994; Aunan *et al.*, 2000; Kuik *et al.*, 2000; Wang and Mauzerall, 2004]. Although not process-based or ecosystem-dependent, such models may be extrapolated to entire ecosystems.

[5] Process-based models allow plant growth responses to vary with dynamic environments, such as high O<sub>3</sub> concentration, elevated CO<sub>2</sub> concentration, and climate change [Tian *et al.*, 1998a]. Several process-based models have attempted to study the effects of O<sub>3</sub> on vegetation [e.g., Reich, 1987; Ollinger *et al.*, 1997; Ollinger, 2002; Martin *et al.*, 2001; Felzer *et al.*, 2004, 2005]. Reich's [1987] model is not actually a process-based model, but he generalized a linear model to describe the response of crops and trees to O<sub>3</sub> and argued that crops were more sensitive to O<sub>3</sub>. Ollinger *et al.* [1997] used O<sub>3</sub>-response relationships with the PnET-II model to simulate tree growth and ecosystem functions. These models can apply the dynamic O<sub>3</sub> damage mechanisms in seedling and mature trees from leaf level to canopy level. Ollinger and his colleagues [Ollinger, 2002] applied the model to study the effect of O<sub>3</sub> on NPP for specific sites within the northeastern U.S. (a reduction in NPP of between 3 and 16%) and the combined effects of CO<sub>2</sub>, O<sub>3</sub>, and N deposition along with the context of historical land use changes for hardwoods in the northeastern U.S. with a new version of PnET (PnET-CN). Felzer *et al.* [2004, 2005] incorporated the algorithms from Reich [1987] and Ollinger *et al.* [1997] for hardwoods, conifers, and crops into a biogeochemical model (i.e., TEM). Their study across the conterminous U.S. indicated a 2.6–6.8% mean reduction in annual NPP in the US during the late 1980s and early 1990s. Unlike Ollinger's and Felzer's work, in which the effects of ozone on stomatal conductance were not considered, Martin *et al.* [2001] incorporated O<sub>3</sub> effects on photosynthesis and stomatal conductance into the functional-structural tree growth model ECOPHYS (<http://www.nrri.umn.edu/default>) by using O<sub>3</sub> flux data. Not only did they combine the well-accepted equations from mech-

anistic biochemical models for photosynthesis (e.g., equations from Farquhar *et al.* [1980] and von Caemmerer and Farquhar [1981]) and the equations from phenological models for stomatal conductance (Ball *et al.* [1987], adapted by Harley *et al.* [1992]), but also explored the underlying mechanisms of O<sub>3</sub>-inhibited photosynthesis models. They found that O<sub>3</sub> damage could reduce both protective scavenging detoxification system (*V<sub>c</sub> max*) and light-saturated rate of electron transport (*J max*) by the accumulated amounts of O<sub>3</sub> above the threshold of damage entering the inner leaves. Considering the advantages and disadvantages of different models in simulating O<sub>3</sub> effects, a coupled mechanistic model that fully couples energy, carbon, nitrogen, and water, as well as vegetation dynamics is needed in the near future [Tian *et al.*, 1998a].

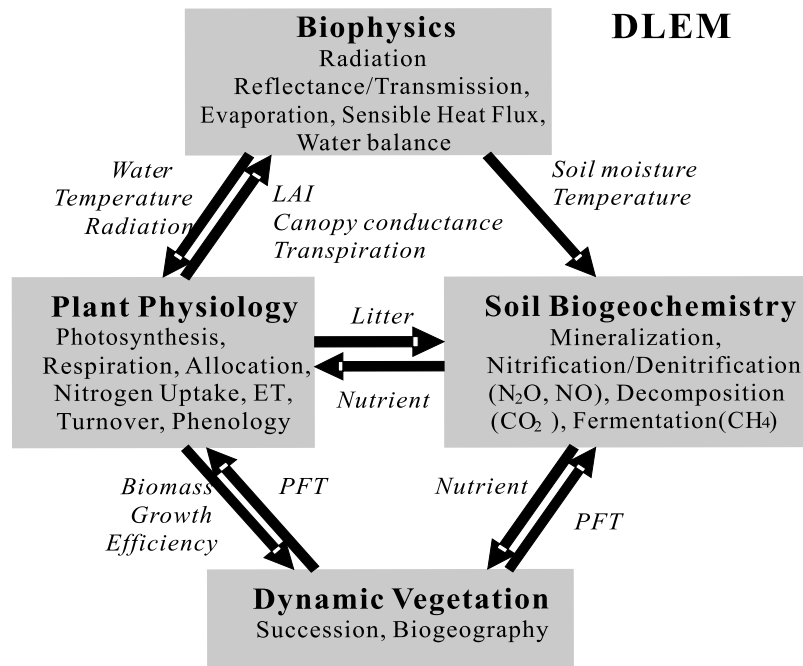
[6] In this research, we used a highly integrated process-based model called Dynamic Land Ecosystem Model (DLEM) (detail description of this model was given by Tian *et al.* [2005]). The dynamic O<sub>3</sub> damage mechanisms were extrapolated from a small spatial scale (leaf level) and a short-term scale into the corresponding long-term mechanism at the ecosystem scale. The O<sub>3</sub> module was primarily based on the work of Ollinger *et al.* [1997]. The equations from Farquhar *et al.* [1980] and Ball *et al.* [1987] were used to simulate photosynthesis and stomatal conductance, similar to Martin *et al.* [2001]. This module simulated O<sub>3</sub> damage on plant photosynthesis and NPP. We also developed the spatial data sets including historical climate, soil information, and land use change across China over a long period. The ozone sensitivities for different PFTs including crops, coniferous trees, hardwoods and other vegetation types, were based on the Reich's compilation of OTC experiments in the U.S., which we assume to be applicable to China as well.

[7] More ozone pollution in China is closely related to domestic food security and the global environment in the future [e.g., Chameides *et al.*, 1999; Akimoto, 2003]. Unlike other studies in China [Aunan *et al.*, 2000; Wang and Mauzerall, 2004; Felzer *et al.*, 2005], we try to illustrate the effects of tropospheric O<sub>3</sub> pollution on terrestrial ecosystem productivity throughout the country between 1961 and 2000. We focus on the analysis of ozone effects on NPP and carbon storage in the context of multiple environmental stresses including increasing O<sub>3</sub>, changing climate, elevated CO<sub>2</sub>, and land use changes (including nitrogen fertilization and irrigation on croplands) across China. In this paper, we first briefly describe our model development, data preparation, and the experimental design, and then examine the relative effects of ozone and other environmental factors on carbon storage across the country. The sensitivity of different PFTs to ozone pollution is also examined. Finally, we discuss and analyze the simulation results and their uncertainty.

## 2. Methods

### 2.1. Dynamic Land Ecosystem Model (DLEM)

[8] The DLEM couples major biogeochemical cycles, hydrological cycle, and vegetation dynamics to generate daily, spatially explicit estimates of water, carbon (CO<sub>2</sub>, CH<sub>4</sub>), and nitrogen fluxes (N<sub>2</sub>O) and pool sizes (C and N) in terrestrial ecosystems (see Figure 1). DLEM includes five



**Figure 1.** Framework of the Dynamic Land Ecosystem Model (DLEM). The DLEM model includes five core components: (1) biophysics, (2) plant physiology, (3) soil biogeochemistry, (4) dynamic vegetation and (5) land use and management [Tian *et al.*, 2005].

core components: (1) biophysics, (2) plant physiology, (3) soil biogeochemistry, (4) dynamic vegetation, and (5) land use and management. The biophysical component includes the instantaneous exchanges of energy, water, and momentum with the atmosphere. It includes aspects of micrometeorology, canopy physiology, soil physics, radiative transfer, hydrology, surface fluxes of energy, moisture, and momentum influences on simulated surface climate. The component of plant physiology in DLEM simulates major physiologic processes, such as photosynthesis, autotrophic respiration, allocation among various parts (root, stem, and leaf), turnover of living biomass, nitrogen uptake and fixation, transpiration, phenology, etc. The component of soil biogeochemistry simulates N mineralization, nitrification/denitrification [Li *et al.*, 2000],  $\text{NH}_3$  volatilization, leaching of soil mineral N, decomposition and fermentation [Huang *et al.*, 1998]. Thus DLEM is able to simultaneously estimate emissions of multiple trace gases ( $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$ ) from soils. The dynamic vegetation component in DLEM simulates two kinds of processes: the biogeographical redistribution when climate changes, and the plant competition and succession during vegetation recovery after disturbances. Like most DGVMs (Dynamic Global Vegetation Models), DLEM builds on the concept of PFT to describe vegetation distributions (Figure 2). The DLEM has also emphasized the simulation of managed ecosystems, including agricultural ecosystems, plantation forests, and pastures. The DLEM 1.0 version has been used to simulate the effects of climate variability and change, atmospheric  $\text{CO}_2$ , tropospheric ozone, land use change, nitrogen deposition, and disturbances (e.g., fire, harvest, hurricanes) on terrestrial carbon storage and fluxes in China [Tian *et al.*, 2005; Chen *et al.*, 2006; Ren *et al.*, 2007]. This model has been calibrated against field data from various ecosystems

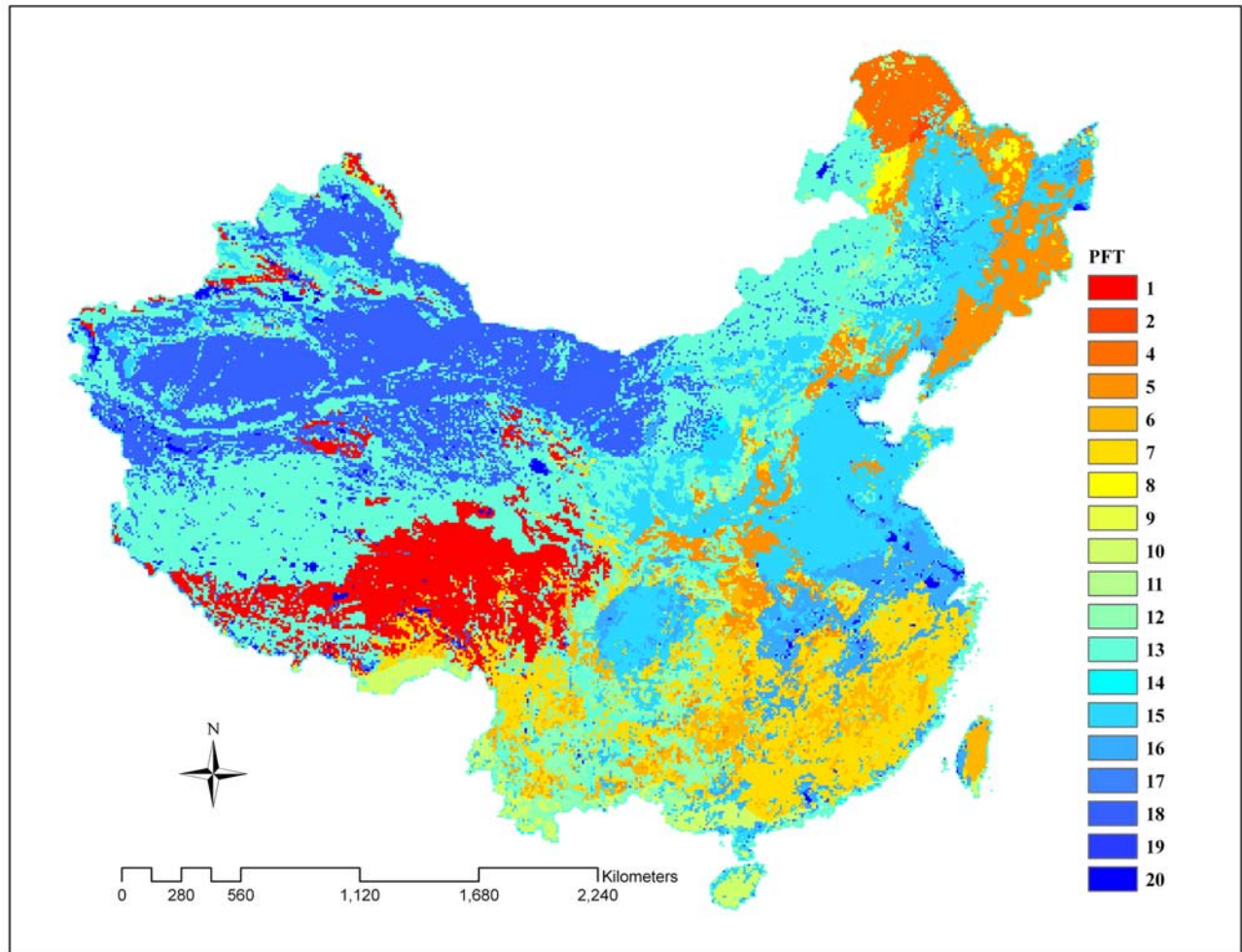
including forests, grassland, and croplands. The simulated results with DLEM have also been evaluated against independent field data [Tian *et al.*, 2005].

[9] In DLEM, the carbon balance of vegetation is determined by the photosynthesis, autotrophic respiration, litter-fall (related to tissue turnover rate and leaf phenology), and plant mortality rate. Plants assimilate carbon by photosynthesis, and use this carbon to compensate for the carbon loss through maintenance respiration, tissue turnover, and reproduction. The photosynthesis module of DLEM estimates the net C assimilation rate, leaf daytime maintenance respiration rate, and gross primary productivity (GPP, unit:  $\text{g C/m}^2/\text{day}$ ). The photosynthesis rate is first calculated on the leaf level. The results are then multiplied by leaf area index to scale up to canopy level [Tian *et al.*, 2005; Chen *et al.*, 2006; Ren *et al.*, 2007; Zhang *et al.*, 2007]. 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:

$$GPP_i = (A_i + Rd_i) \times LAI_i \times \text{dayl} \quad (1)$$

$$A_i = f(PPFD_{\text{leaf}i}, g_i, \text{leaf}N_i, T_{\text{day}}, Ca, \text{dayl}) \quad (2)$$

where  $GPP$  ( $\text{gC/m}^2/\text{day}$ ) is the gross primary productivity of ecosystems for leaf type  $i$ ;  $i$  is leaf type (sunlit leaf or shaded leaf);  $A$  ( $\text{g/s/m}^2$  leaf) and  $Rd$  ( $\text{g/s/m}^2$  leaf) are daytime photosynthesis rate and leaf respiration rate respectively;  $LAI$  is leaf area index;  $\text{dayl}$  (s) is the length of daytime;  $PPFD$  ( $\mu\text{mol/m}^2/\text{s}$ ) is the photosynthetic photon flux density;  $g$  (m/s) is the stomatal conductance of leaf to  $\text{CO}_2$  flux;  $T_{\text{day}}$  ( $^\circ\text{C}$ ) is daytime temperature;  $Ca$  (ppmv) is



**Figure 2.** Contemporary plant functional types in China used in DLEM. 1, tundra; 2, boreal broadleaf deciduous forest; 4, boreal needleleaf deciduous forest; 5, temperate broadleaf deciduous forest; 6, temperate broadleaf evergreen forest; 7, temperate needleleaf evergreen forest; 8, temperate needleleaf deciduous forest; 9, tropical broadleaf deciduous forest; 10, tropical broadleaf evergreen forest; 11, deciduous shrub; 12, evergreen shrub; 13, C3 grass; 14, C4 grass; 15, dry farmland; 16, paddy land; 17, wetland; 18, Gebi and desert; 19, build-up area; 20, water body.

the atmospheric  $\text{CO}_2$  concentration;  $leafN$  ( $\text{gN/m}^2$  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 photosynthesis rate. In this study, NPP in an ecosystem and annual net carbon exchange ( $NCE$ ) of the terrestrial ecosystem with the atmosphere were computed with following equations:

$$NPP = GPP - R_d \quad (3)$$

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

where NPP is the net primary productivity,  $R_d$  is the plant respiration,  $R_H$  is soil respiration,  $E_{NAD}$  is the magnitude of the carbon loss from a natural disturbance and is assigned as 0 here because of the difficulty of being simulated at present conditions,  $E_{AD}$  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 [McGuire *et al.*, 2001; Tian *et al.*, 2003]. For natural ecosystems,  $E_P$  and  $E_{AD}$  are equal to 0, and so  $NCE$  is equal to net ecosystem production ( $NEP$ ). Unlike the other models which estimate the cropland C cycle on the basis of the simulation of potential vegetation of the agricultural grids [McGuire *et al.*, 2001], the agricultural ecosystems in DLEM are not based on natural vegetation, but parameterized against several intensively studied agricultural sites in China (<http://www.cerndata.ac.cn/>).

[10] To simulate the detrimental effect of air pollution on ecosystem productivity, an ozone module was developed on the basis of previous work [Ollinger *et al.*, 1997; Felzer *et al.*, 2004, 2005], in which the direct effect of ozone on photosynthesis and indirect effect on stomatal conductance by changing intercellular  $\text{CO}_2$  concentration were simulated. Here the ratio of ozone damage to photosynthesis is defined as  $O_{3\text{eff}}$ , similar to Ollinger *et al.* [1997], and the sensitivity coefficient  $a$  for each different plant functional type (Table 1)

**Table 1.** Values of Sensitivity Coefficient  $\alpha$  for Different Functional Types<sup>a</sup>

Functional Types	$\alpha$ Coefficient	Reference
Crops	$3.9 \times 10^{-6}$ ( $\delta = 5.27 \times 10^{-7}$ )	Reich [1987]
Coniferous trees	$0.7 \times 10^{-6}$ ( $\delta = 2.45 \times 10^{-7}$ )	Reich [1987]
Deciduous trees and other vegetation types	$2.6 \times 10^{-6}$ ( $\delta = 2.3 \times 10^{-7}$ )	Ollinger et al. [1997]

<sup>a</sup>The values of  $\alpha$  directly modified from Felzer et al. [2004].

is based on the work of Felzer et al. [2004]. The range of  $\alpha$  is  $2.6 \cdot 10^{-6} \pm 2.8 \cdot 10^{-7}$  for hardwoods (based on the value used by Ollinger et al. [1997]),  $0.8 \cdot 10^{-6} \pm 3.6 \cdot 10^{-7}$  for conifers (based on pines), and  $4.9 \cdot 10^{-6} \pm 1.6 \cdot 10^{-7}$  for crops which was calculated from the empirical model of Reich [1987]. The errors are based on the standard deviation of the slope from the dose response curves and the standard error of the mean stomata conductance.

$$GPP_{O_3} = GPP \times O_{3eff} \quad (5)$$

$$O_{3eff} = 1 - (\alpha \times g_s \times AOT_{40}) \quad (6)$$

$$g_s = f(GPP_{O_3}) \quad (7)$$

Here,  $GPP_{O_3}$  is limited  $GPP$  because of ozone effect;  $g_s$  is the stomatal conductance ( $\text{mms}^{-1}$ );  $AOT_{40}$  is a cumulative ozone index (the accumulated hourly ozone dose over a threshold of 40 ppb in ppb/h), and in this study we use a monthly accumulative index as in the work by Felzer et al. [2004]. The AOT40 index has often been used to represent vegetation damage due to ozone [Fuhrer et al., 1997]. Because of limited ozone data throughout China, we use the model-developed AOT40 values from Felzer et al. [2005].

[11] Our photosynthesis module, based on Farquhar et al.'s [1980] model, has the potential ability to use ozone concentration as input, similar to Martin et al. [2001], if the ozone flux data are available in the future. In DLEM, the leaf C: N ratio is also affected by ozone. We do not use this mechanism in the current study because of the ambiguous role of ozone on plant C:N ratio [Lindroth et al., 2001].

## 2.2. Input Data

[12] Input data sets include (1) elevation, slope, and aspect maps which are derived from 1 km resolution digital elevation data set of China (<http://www.wdc.cn/wdcdrre>); (2) soil data sets (pH, bulk density, depth to bedrock, soil texture represented as the percentage content of loam, sand and silt) which are derived from the 1:1 million soil map based on the second national soil survey of China [Wang et al., 2003; Shi et al., 2004; Zhang et al., 2005; Tian et al., 2006]; (3) vegetation map (or land cover map) from the 2000 land use map of China (LUCC\_2000) which was developed from Landsat Enhanced Thematic Mapper (ETM) imagery [Liu et al., 2005a]; (4) potential vegetation map, which is constructed by replacing the croplands of LUCC 2000 with potential vegetation in global potential vegetation maps developed by Ramankutty and Foley [1998]; (5) standard IPCC (Intergovernmental Panel on

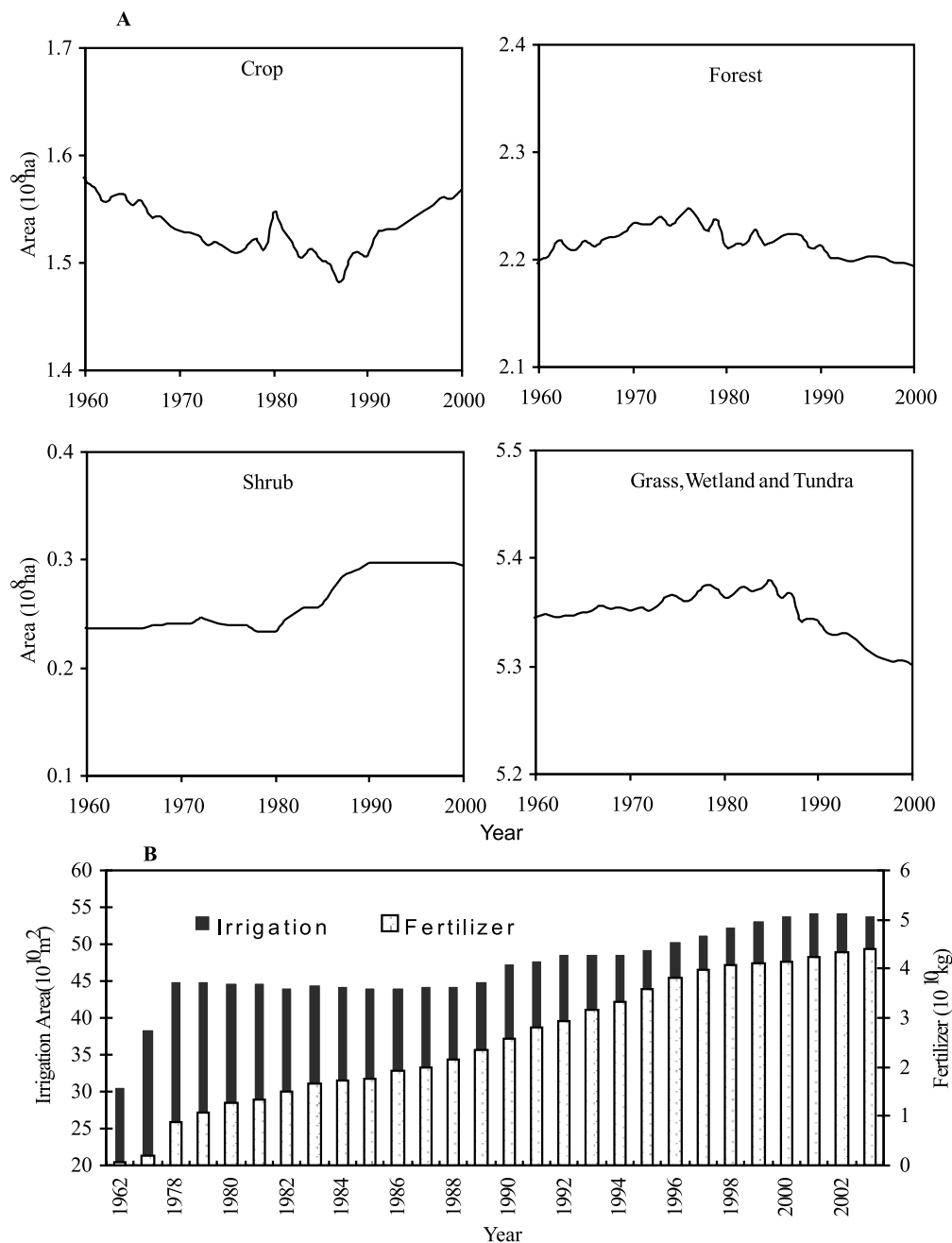
Climate Change) historical CO<sub>2</sub> concentration data set [Enting et al., 1994]; (6) AOT40 data set (see below for detail information); (7) long-term land use history (cropland and urban distribution of China from 1661 to 2000) which is developed on the basis of three recent (1990, 1995 and 2000) land cover maps [Liu et al., 2003, 2005a, 2005b] and historical census data sets of China [Ge et al., 2003; Xu, 1983]; and (8) daily climate data (maximum, minimum, and average temperature, precipitation, and relative humidity). Seven hundred and forty six climate stations in China and 29 stations from surrounding countries were used to produce daily climate data for the time period from 1961 to 2000, using an interpolation method similar to that used by Thornton et al. [1997]. To account for cropland management, we also used data from the National Bureau of Statistics of China, which recorded annual irrigation areas and fertilizer amounts in each province from 1978 to 2000 (Figure 3b). We did not construct an irrigation data set because of lack of data. We simulated the effects of irrigation by refilling the soil water pool to field capacity whenever cropland soil reached wilt point. All data sets have a spatial resolution of  $0.5^\circ \times 0.5^\circ$ , and Climate and AOT40 data sets have been developed on daily time step while CO<sub>2</sub> and land use data sets on yearly time step.

### 2.2.1. Description of Ozone Data

[13] The methods used for monitoring ozone vary among the limited ground ozone monitoring sites in China [Chameides et al., 1999; Li et al., 2000; Chen et al., 1998]. Therefore it is difficult to spatially develop a historical AOT40 data set based on the interpolation of site-level data like Felzer et al. [2004] for the U.S. In this study, the AOT40 data set was derived from the global historical AOT40 data sets constructed by Felzer et al. [2005]. This AOT40 index is calculated from combining geographic data from the MATCH model (Multiscale Atmospheric Transport and Chemistry) [Lawrence and Crutzen, 1999; Rasch et al., 1997; von Kuhlmann et al., 2003] with hourly zonal ozone from the MIT IGSM (Integrated Global Systems Model). The average monthly boundary layer MATCH ozone values for 1998 are scaled by the ratio of the zonal average ozone from the IGSM (Integrated Global Systems Model), which are 3-hourly values that have been linearly interpolated to hourly values, to the zonal ozone from the monthly MATCH to maintain the zonal ozone values from the IGSM [Wang et al., 1998; Mayer et al., 2000]. This procedure was done for the period 1977–2000. From 1860 to 1976, the zonal ozone values were assumed to increase by 1.6% per year on the basis of Marenco et al. [1994].

[14] The AOT40 (Figure 4) shows significant increase of ozone pollution in the past 40 a, and the trend accelerated rapidly since the early 1990s, possibly because of the rapid urbanization during that period in China [Liu et al., 2005b]. The data set shows seasonal variation of AOT40, with the first peak of ozone concentration occurring in early summer and the second in September. Both peaks appear approximately at the critical time (the growth and harvest seasons) for crops in China. Thus ozone pollution may have significant impacts on crop production in China.

[15] Although the AOT40 generally increased throughout the nation, the severity of ozone pollution varied from region-to-region and from season-to-season (Figure 5). The central-eastern section of north China experienced



**Figure 3.** (a) Variations of land use and (b) irrigation area ( $10^{10}$  m<sup>2</sup>) and fertilizing amount ( $10^{10}$  kg).

severe ozone pollution, especially in spring and summer. The greatest increase of AOT40 appeared in winter of northwest China, probably due to the rapid industrialization and the transport of air pollution from Europe [Akimoto, 2003]. In contrast, the change of AOT40 in south China is relatively low despite the large urban population and rapid industrial development in this region.

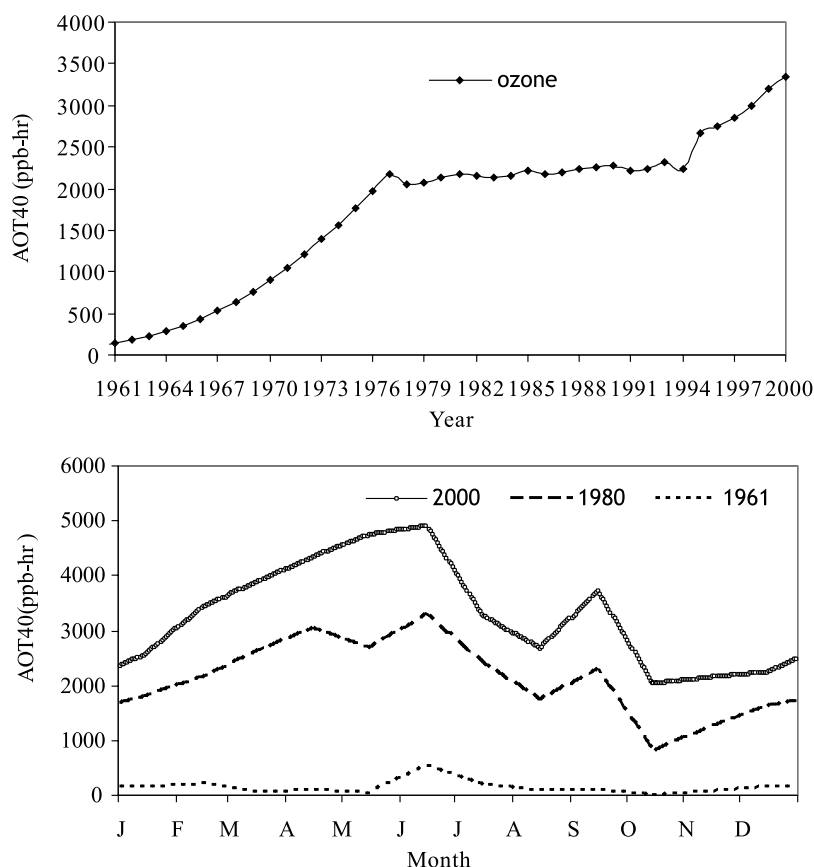
### 2.2.2. Description of Other Input Data

[16] From 1961 to 2000, the CO<sub>2</sub> concentration steadily increased from 312 ppmv to 372 ppmv (Figure 6a), while temperature and precipitation fluctuated substantially (Figures 6b and 6c). Since the mid-1980s, China experienced an observable climate warming. The annual precip-

itation in the 1990s was higher than that in the 1980s. There was a relatively long dry period between 1965 and 1982, except for high annual precipitation in 1970 and 1974 (Figures 6c and 6e). Figure 3a shows that since the late 1980s, cropland expanded, while forestry and other land areas gradually decreased.

### 2.3. Simulation Design

[17] In this study, six experiments were designed to analyze the effects of ozone on NPP, NCE, and carbon storage in terrestrial ecosystems of China (Table 2). Experiment I was used to examine the impact of transient ozone on terrestrial ecosystem productivity while holding other environmental factors constant. Experiments II and III were



**Figure 4.** (a) Annual monthly AOT40 (ppb/h) mean from 1961 to 2000 and (b) monthly AOT40 (ppb/h) in 1961, 1980 and 2000. Note: From atmospheric chemistry model, MATCH (Multiscale Atmospheric Transport and Chemistry) [Lawrence and Crutzen, 1999; Mahowald et al., 1997; Rasch et al., 1997; von Kuhlmann et al., 2003] and IGSM (Integrated Global Systems Model) [Wang et al., 1998; Wang and Prinn, 1999; Mayer et al., 2000].

used to analyze the combined effects of  $O_3$  and  $CO_2$  fertilization and of  $O_3$  and climate change. Both experiments can help better determine the relative impacts of  $O_3$ ,  $CO_2$  and climate on the ecosystem. Experiments IV simulated the overall effect of climate change, atmospheric change, and land use change. Experiment V was set up to study the overall combined effect without irrigation. The final experiment VI without ozone effects is used for comparison against the other experiments.

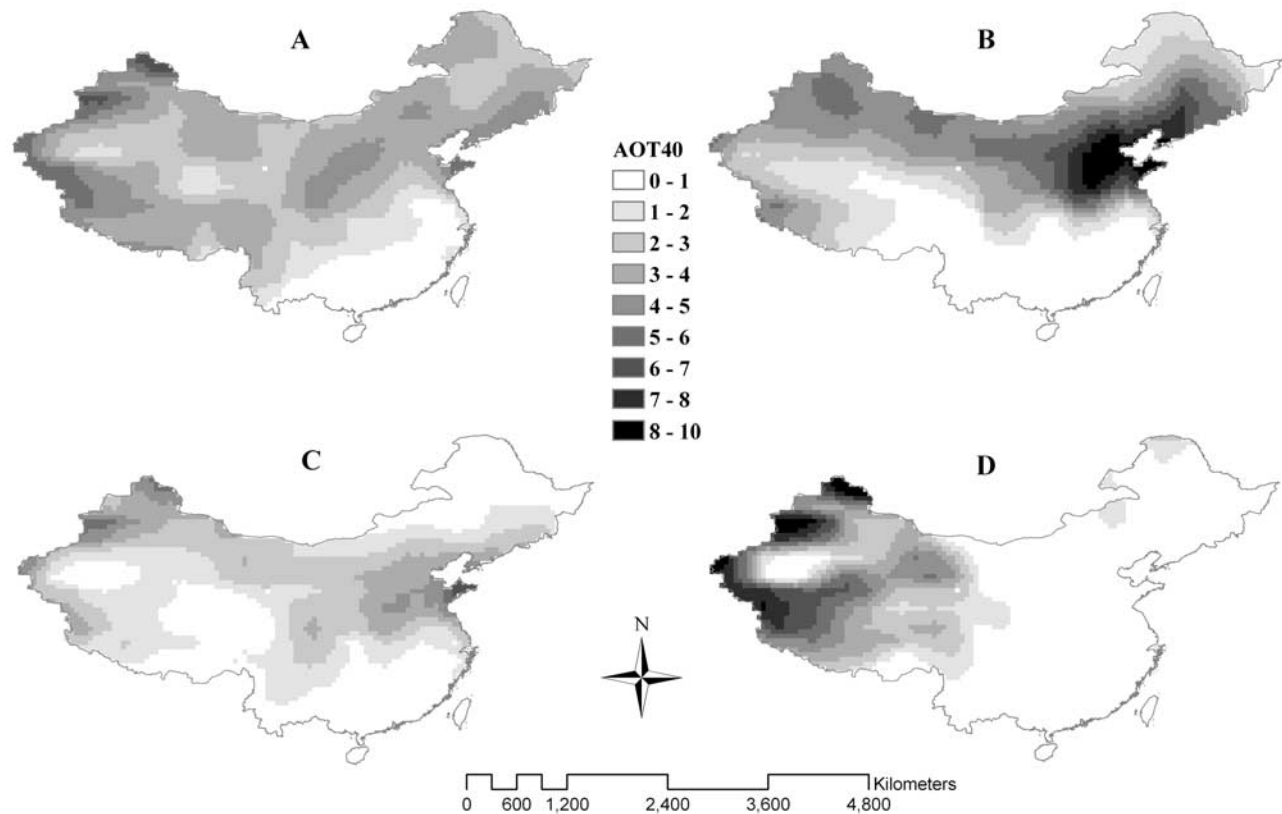
[18] The model simulation began with an equilibration run to develop the baseline C, N, and water pools for each grid. A spin-up of about 100 a was then applied if the climate change was included in the simulation scenario. Finally, the model ran in transient mode driven by the daily or/and annual input data.

### 3. Results and Analyses

#### 3.1. Overall Change in Net Primary Productivity and Carbon Storage

[19] In the simulation experiments, there were negative effects of  $O_3$  on average NPP and carbon storage during the study period (1961–2000). Average annual NPP and total C storage from the 1960s to 1990s in China increased by 0.66% and 0.06%, respectively, under the full factorial

(climate, land use,  $CO_2$ , and  $O_3$  were changed, hereafter referred to as OCLC), while they increased by 7.77% and 1.63%, respectively, under the scenario without  $O_3$  (hereafter CLC) (Table 3). This difference indicates that under the full factorial,  $O_3$  decreased NPP (about  $-1.64\%$  in 1960s and  $-8.11\%$  in 1990s) and total C storage (about  $-0.06\%$  in 1960s and  $-1.61\%$  in 1990s) in China's terrestrial ecosystems. Although NPP and total C storage in both scenarios increased over time, the soil and litter C storage decreased ( $-0.18\%$  and  $-0.67\%$ , respectively) under the full factorial, while they increased by 0.30% and 1.63%, respectively, under the scenario without  $O_3$ . Therefore  $O_3$  reduced soil and litter C storage by about 0.03% and 0.16% in the 1960s and 0.52% and 1.84% in the 1990s, respectively, in China. The model results show that NPP and carbon storage, including vegetation carbon, soil carbon, and litter carbon, decreased with  $O_3$  exposure, and the reduced NPP was more than the decrease in carbon storage. The changing rates in the 1960s and 1990s indicate that increasing  $O_3$  concentrations could result in less NPP and carbon storage, which further implies that under the influence of  $O_3$  alone, China's soil ecosystem would be a net C source, while without  $O_3$ , it would have been a net C sink (Table 3).



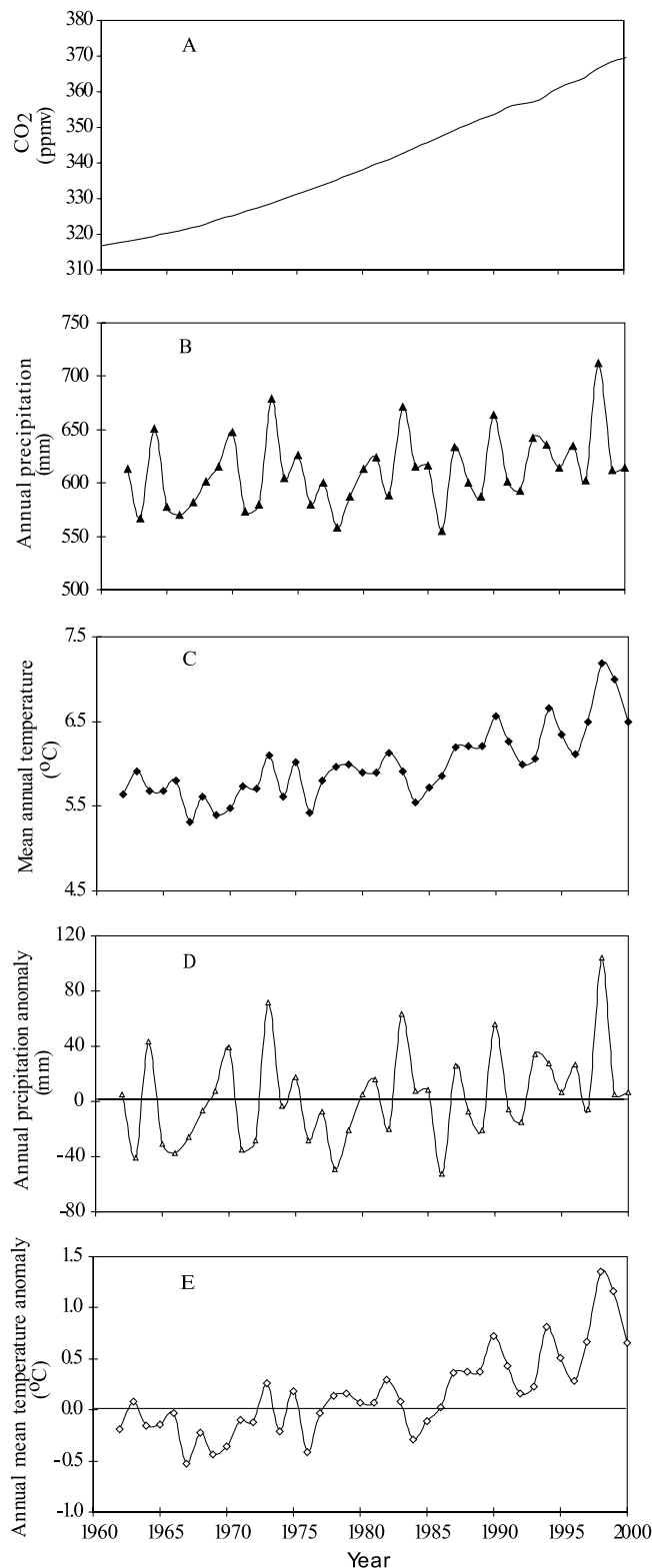
**Figure 5.** Average monthly AOT<sub>40</sub> in (a) spring, (b) summer, (c) autumn, and (d) winter from 1990 to 2000 in China (unit is 1000 ppb/h or ppm/h). Note: From atmospheric chemistry model, MATCH (Multiscale Atmospheric Transport and Chemistry) [Lawrence and Crutzen, 1999; Mahowald et al., 1997; Rasch et al., 1997; von Kuhlmann et al., 2003] and IGSM (Integrated Global Systems Model) [Wang et al., 1998; Wang and Prinn, 1999; Mayer et al., 2000].

[20] The results of the accumulated NCE across China under three simulation experiments, including O<sub>3</sub>-only, combined effects without O<sub>3</sub> (CLC) and with O<sub>3</sub> (OCLC) from 1961 to 2000, indicate that O<sub>3</sub> effects could cause carbon release from the terrestrial ecosystem to the atmosphere (Table 4). The accumulated NCE was  $-919.1$  Tg C under the influence of O<sub>3</sub> only,  $1,177.4$  Tg C under the combined influence of changed climate, CO<sub>2</sub> and land use (CLC), and  $677.3$  Tg C under the full factorial (OCLC). These values imply that China was a CO<sub>2</sub> source when influenced only by O<sub>3</sub>, but it was a sink under the influence of both CLC and OCLC scenarios. The accumulated NCE influenced by OCLC was  $620.4$  Tg C less (OCLC-CLC) than that influenced by CLC, implying that the interactions between O<sub>3</sub> and other factors (CO<sub>2</sub>, climate, or land use change) were very strong. These interactions decreased the emissions of CO<sub>2</sub> from terrestrial ecosystems. For the 1990s, a period with rapid atmospheric O<sub>3</sub> change, our simulated results show that the cumulative NCE under the full factorial (OCLC) throughout China decreased, compared to the results without O<sub>3</sub> influence (CLC) (Table 3 and Figure 7). In the central-eastern China and northeastern China, some places even released  $150$  g/m<sup>2</sup> more C into the atmosphere under O<sub>3</sub> influences during the 1990s.

[21] Accumulated carbon storage of different PFTs under the three simulation experiments indicate that PFTs respond

very differently to increasing O<sub>3</sub> concentration and its interaction with other environmental factors (Table 4). From 1961 to 2000, the accumulated NCE for different PFTs with O<sub>3</sub> exposure decreased by only  $0.1$  Tg C in wetlands up to  $615.9$  Tg C in temperate broadleaf deciduous forests. Accumulated NCE under influences of CLC for different PFTs ranged from a decrease of  $67.7$  Tg C (Tundra) to an increase of  $720.4$  Tg C (temperate needleleaf evergreen forest), while accumulated NCE under influences of OCLC ranged from a decrease of  $547.0$  Tg C (temperate broadleaf deciduous forest) to an increase of  $720.4$  Tg C (temperate needleleaf evergreen forest). This range implies that temperate broadleaf deciduous forest was the biggest C source under the full factorial and that temperate needleleaf evergreen forest was the biggest C sink from 1961 to 2000. Compared with the combined effect with O<sub>3</sub> (OCLC) and without O<sub>3</sub> (CLC), the O<sub>3</sub>-only scenario releases more C for all different PFTs. Compared with the combined effect without O<sub>3</sub> (CLC), the combined effect with O<sub>3</sub> (OCLC) resulted in less accumulated NCE for temperate broadleaf deciduous forest ( $321.2$  Tg C) and dry farmland crops ( $46.5$  Tg C) than other PFTs, which means that temperate broadleaf deciduous forest and dry farmland were more sensitive to O<sub>3</sub> than other PFTs. In general, we found that C<sub>3</sub> grass was more sensitive than C<sub>4</sub> grass to O<sub>3</sub>, dry farmland





**Figure 6.** Variations in (a) mean annual atmospheric CO<sub>2</sub> concentration, (b) mean annual temperature, (c) annual precipitation, (d) annual precipitation anomalies, and (e) annual temperature anomalies (relative to 1961–1990 normal period) from 1961 to 2000.

was more sensitive than paddy farmland, and deciduous forest was more sensitive than needleleaf forest.

[22] Overall results indicate that O<sub>3</sub> has negative impacts on terrestrial ecosystem production (Figure 4 and Table 3), and the negative effects become severe because the O<sub>3</sub> concentration increased across China in the past decades, especially after the 1990s [e.g., *Aunan et al.*, 2000]. Biomes had complicated responses of carbon storage to O<sub>3</sub> because of the different sensitivities of each PFT as well as different environmental conditions. For example, some arid sites exhibit small ozone effect on photosynthesis because low stomatal conductance in arid plants leads to relatively low ozone uptake. Some other studies showed that O<sub>3</sub>-induced reductions in photosynthesis were accompanied by decreased water use efficiency (WUE), however, resulting in even larger reductions in productivity, particularly at arid sites [Ollinger *et al.*, 1997]. This fact may be the reason that wetlands show relatively low reduction in carbon storage (Table 4), and dry farmland crops are more sensitive to O<sub>3</sub> than paddy farmland crops. This might be also because parameters of crops in the Reich model [Reich, 1987] are more sensitive to O<sub>3</sub> than those in deciduous and coniferous forests. In addition, in response to elevated O<sub>3</sub> concentration, plants allocate more carbon to leaves and stems than roots because of increased defense mechanisms [e.g., *Younglove et al.*, 1994; *Piikki et al.*, 2004]. This effect may result in higher carbon storage loss in broadleaf deciduous forests than in evergreen forests. It is clearly needed to address variations in biome-level responses to ozone pollution.

### 3.2. Spatiotemporal Variations of C Flux and C Storage

[23] Mean annual NPP changes from the 1960s to the 1990s under the O<sub>3</sub>-only scenario showed a significant spatial pattern (Figure 8). The mean annual NPP decreased the most in the eastern China partially because the eastern China has experienced faster development in urbanization, industrialization, and agricultural intensification in the past several decades, than remote areas in the western China [Aunan *et al.*, 2000; Wang and Mauzerall, 2004; Liu *et al.*, 2005a, 2005b]. This development is closely related to an increased use of fossil fuels and fertilizer. The imbalance of regional O<sub>3</sub> concentrations could also result in fluctuations of annual NPP.

[24] Under the influence of O<sub>3</sub> and CO<sub>2</sub>, the simulation results illustrate that mean annual NPP in the 1990s increased by 140.6 Tg C compared to the 1960s (Figures 8 and 9); the total carbon storage increased by 46.3Tg over the past 40 a because of the accumulative increase in vegetation and soil C storage (Figure 10). The increased C storage may be attributed to the direct effects of increasing atmospheric CO<sub>2</sub> [Melillo *et al.*, 1993; Tian *et al.*, 1999, 2000], however, ozone can partially compensate for the positive effects of CO<sub>2</sub> fertilization.

[25] DLEM estimates that the total carbon storage for potential vegetation under O<sub>3</sub> and climate influences decreased by 15.9 Tg C. This decrease could mainly be attributed to the large decrease in soil C storage while vegetation C decreased relatively little from 1961 to 2000 (Figure 10). The interannual variation of NPP had a similar trend with the historical annual precipitation from 1961

**Table 2.** Experimental Arrangement Including CO<sub>2</sub>, Climate, Land Use and Human Being Management (Fertilizer and Irrigation)<sup>a</sup>

	Scenarios	O <sub>3</sub>	Climate	CO <sub>2</sub>	Land Use	Fertilizer	Irrigation
	Balance	0	constant	constant	constant	0	0
I	O <sub>3</sub> only	historical	constant	constant	constant	0	0
II	O <sub>3</sub> _CO <sub>2</sub>	historical	constant	historical	constant	0	0
III	O <sub>3</sub> _Climate	historical	historical	constant	constant	0	0
IV	O <sub>3</sub> _Climate_Lucc_CO <sub>2</sub>	historical	historical	historical	historical	historical	historical
V	O <sub>3</sub> _Climate_Lucc_CO <sub>2</sub> _N	historical	historical	historical	historical	historical	0
VI	Climate_Lucc_CO <sub>2</sub>	0	historical	historical	historical	historical	historical

<sup>a</sup>Climate\_lucc\_CO<sub>2</sub>, CLC; O<sub>3</sub>\_climate\_lucc\_CO<sub>2</sub>, OCLC.

to 2000 (Figures 9 and 6c). For example, annual NPP decreased as less precipitation occurred in the late 1960s and the 1970s (Figure 9). From 1961 to 2000, because of the influence of the monsoon climate, precipitation and temperature in China exhibited large interannual variability during the study period. The results indicate that NPP was more sensitive to changes in precipitation than in temperature, while soil carbon storage was more closely linked to temperature through decomposition responses. In addition, the changing pattern of mean annual NPP from the 1960s to 1990s indicates that NPP in the areas of the eastern and northern China decreased more than other areas over the same time period (Figure 8b). Those variations might be related to the magnitude and spatial distribution of rainfall from seasonal to decadal [Fu and Wen, 1999; Tian et al., 2003]. Therefore the combined effects of changes in air temperature and precipitation with increasing O<sub>3</sub> concentration are complex, and the NPP loss might result from the balance among O<sub>3</sub>, CO<sub>2</sub> and water uptake through changing stomatal conductance due to the combined effects of O<sub>3</sub>, temperature and CO<sub>2</sub>.

[26] The above analyses address the response of potential terrestrial ecosystems to historical O<sub>3</sub> concentration, changing climate, and atmospheric CO<sub>2</sub> concentration. Land use change as well as management, however, has substantially modified land ecosystems across China in the past 40 a [e.g., Liu et al., 2005a, 2005b]. On the basis of the DLEM simulation, the total terrestrial carbon storage in China increased by 16.8 Tg C during 1960–2000 (Figure 10). Annual NPP over time in the OCLC (simulation experiment IV) increased slightly (mean 0.1%), and the variation of interannual NPP is similar to the result from climate change (Figure 9). The distribution of the annual NPP difference between the 1960s and the 1990s in OCLC scenario indicates that the NPP changes were smaller than in the O<sub>3</sub>-only scenario, which could be caused by the modification of the interactive effects of changing climate, increasing CO<sub>2</sub>, and land use change. Compared with the effects of OCLC with irrigation and without irrigation (Figure 9),

there was a mean 3% reduced rate of annual NPP from 1961 to 2000. The result implies that water conditions could alter the ozone-induced damage.

## 4. Discussion

### 4.1. Comparison With Estimates of Ozone Damage From Other Research in China

[27] Research of ozone effects on crop growth and yield loss in China has been conducted since 1990 by using field experiments and model simulations. Field studies have shown that ozone exposure could result in crop yield reductions of 0–86% under different experimental treatments with varying ozone concentrations and duration [e.g., Wang and Guo, 1990; Huang et al., 2004]. Estimates of crop yield loss on the regional scale, such as the Yangtze River Delta and national level, were established [Feng et al., 2003; Wang and Mauzerall, 2004; Aunan et al., 2000], and results indicate crop loss of 0–23% from historical ozone in China and 2.3–64% as ozone rises in the future [Wang et al., 2007]. Felzer et al. [2005] found that crops were more sensitive to ozone damage at low ozone levels both in China and Europe than in the U.S. All the above crop studies and assessments of yield loss were based on empirical exposure-response relationships for crop yield and ozone. However, there are few studies based on process-based ecosystem models to estimate the effects of ozone damage on diverse PFTs on a national level. In our study, the DLEM model was used to address the influence of ozone concentration on NPP and carbon storage across China during the past 40 a from 1960 to 2000, and we estimated the influence of historical ozone on different PFTs. Similar to previous studies, there was a reduction of NPP when considering ozone effects. Also the ozone effects on terrestrial ecosystem across China was consistent with the study of Felzer et al. [2005], that spatial variations were largely due to varied ozone concentration, climate change and other stress factors. The large damage peaked in the eastern China with the greatest reduction in NPP over 70% for some places.

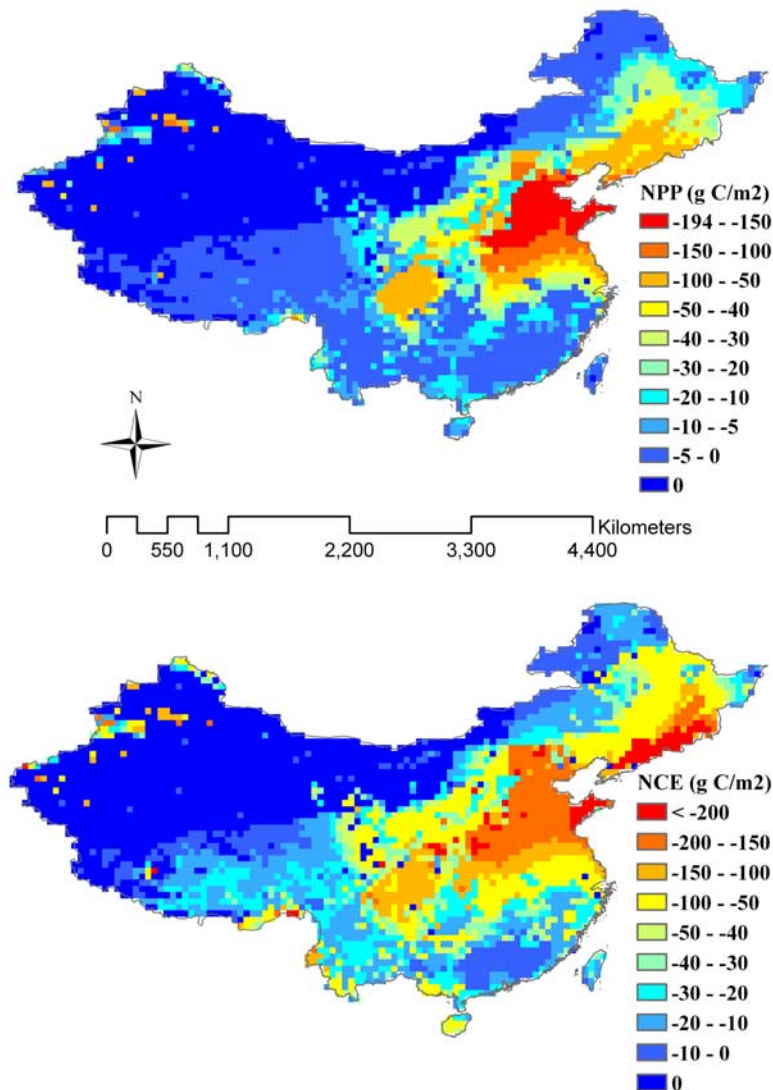
**Table 3.** Overall Changes in Carbon Fluxes and Pools During 1961–2000

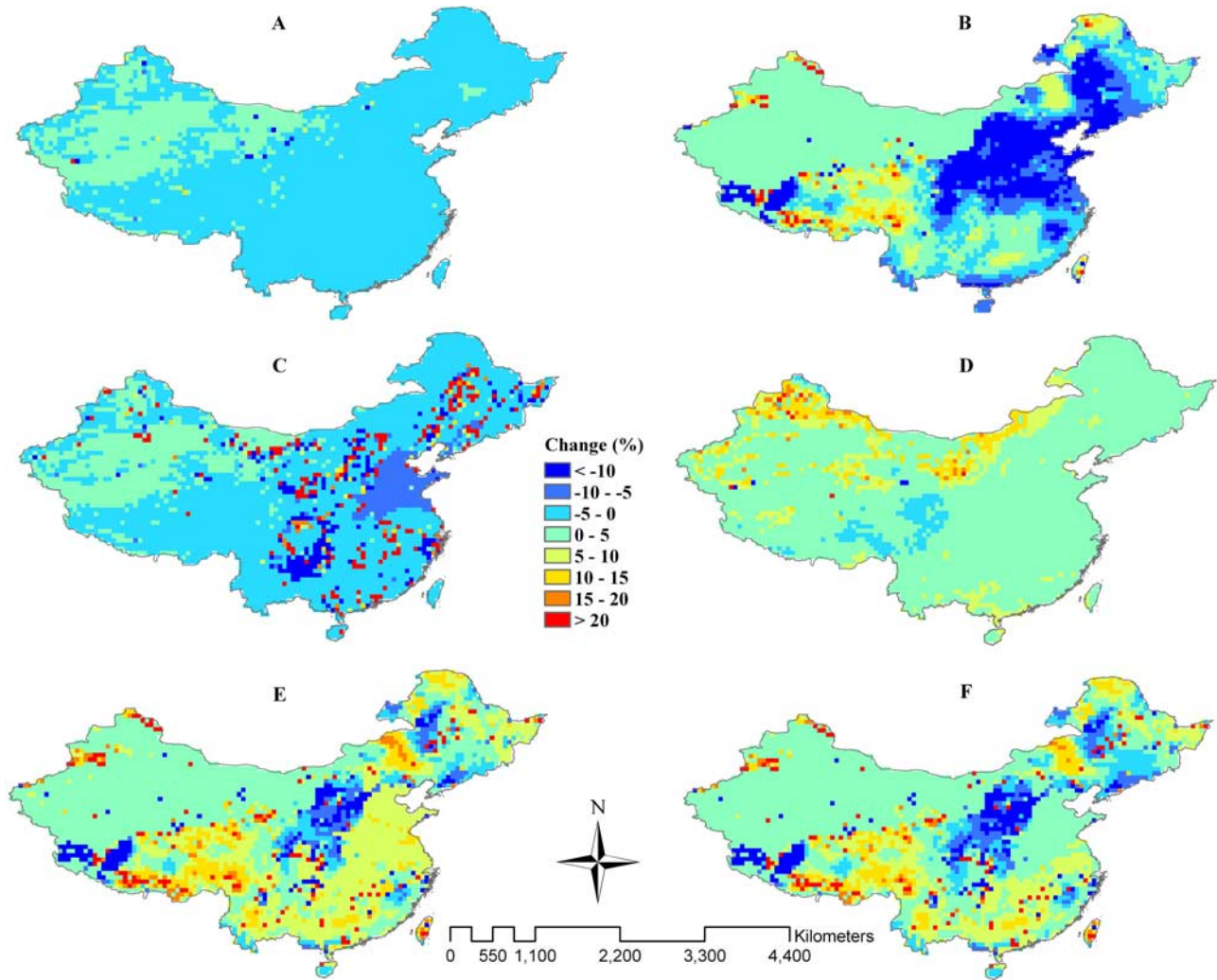
Scenarios	O <sub>3</sub> Climate_Lucc_CO <sub>2</sub>			Climate_Lucc_CO <sub>2</sub>			Difference With O <sub>3</sub> and Without O <sub>3</sub>	
	1960s, Pg	1990s, Pg	Net Change, %	1960s, Pg	1990s, Pg	Net Change, %	1960s, %	1990s, %
NPP	3.04	3.06	0.66	3.09	3.33	7.77	−1.64	−8.11
Veg C	26.02	26.32	1.15	26.03	27.38	5.19	−0.04	−3.87
Soil C	59.79	59.68	−0.18	59.81	59.99	0.30	−0.03	−0.52
Litter C	19.32	19.19	−0.67	19.35	19.55	1.03	−0.16	−1.84
Total C	105.14	105.2	0.06	105.2	106.92	1.63	−0.06	−1.61

**Table 4.** Accumulated NCE of Different Plant Functional Types Under Three Scenarios Including O<sub>3</sub> Only, CLC, OCLC and the Difference Between CLC and OCLC From 1961 to 2000<sup>a</sup>

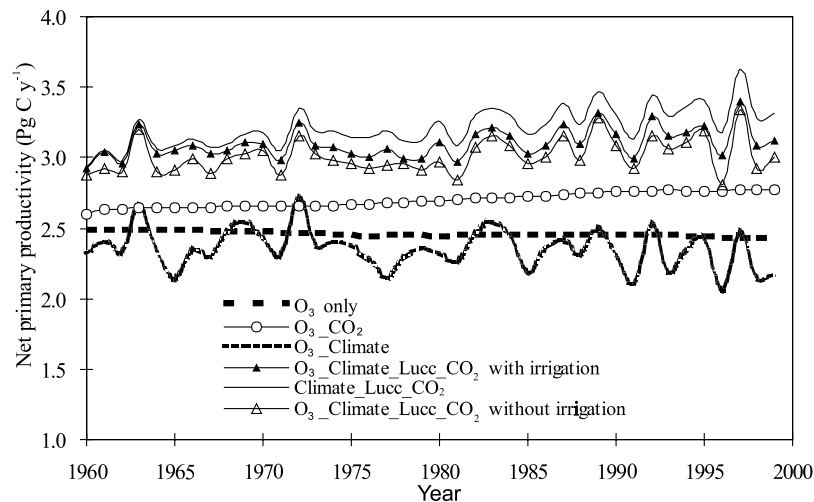
Type Number	Plant Functional Types	Scenarios			
		O <sub>3</sub> Only, Tg C(10 <sup>12</sup> C)	CLC, Tg C(10 <sup>12</sup> C)	OCLC, Tg C(10 <sup>12</sup> C)	OCLC-CLC, Tg C(10 <sup>12</sup> C)
1	tundra	-30.6	-67.7	-130.2	-62.5
2	boreal broadleaf deciduous forest	-1.2	-17	-18.3	-1.3
4	boreal needleleaf deciduous forest	-9.9	94.9	85.1	-9.7
5	temperate broadleaf deciduous forest	-615.9	-225.8	-547.0	-321.3
6	temperate broadleaf evergreen forest	-26	231.8	206.5	-25.3
7	temperate needleleaf evergreen forest	-72.1	720.4	720.4	43.2
8	temperate needleleaf deciduous forest	-5.7	14.9	14.9	-4.0
10	tropical broadleaf evergreen forest	-19.7	-48.1	-48.0	-19.8
11	deciduous shrub	-4.9	17.9	17.9	-5.8
12	evergreen shrub	-11.5	37.1	37.1	13.6
13	C3 grass	-120.6	301.6	301.6	-136.9
14	C4 grass	-0.9	24.4	3.8	-4.0
15	wetland	-0.1	96.7	93.7	-30.1
16	dry farmland		-3.6	-50.1	-46.5
17	paddy farmland		-0.1	-10.1	-10.0
	total accumulated NCE in China since 1961	-919.1	1177.4	677.3	-620.4

<sup>a</sup>There is no dry farmland and paddy farmland under the scenario of O<sub>3</sub> only. Climate\_lucc\_CO<sub>2</sub>, CLC; O<sub>3</sub>\_climate\_lucc\_CO<sub>2</sub>, OCLC.

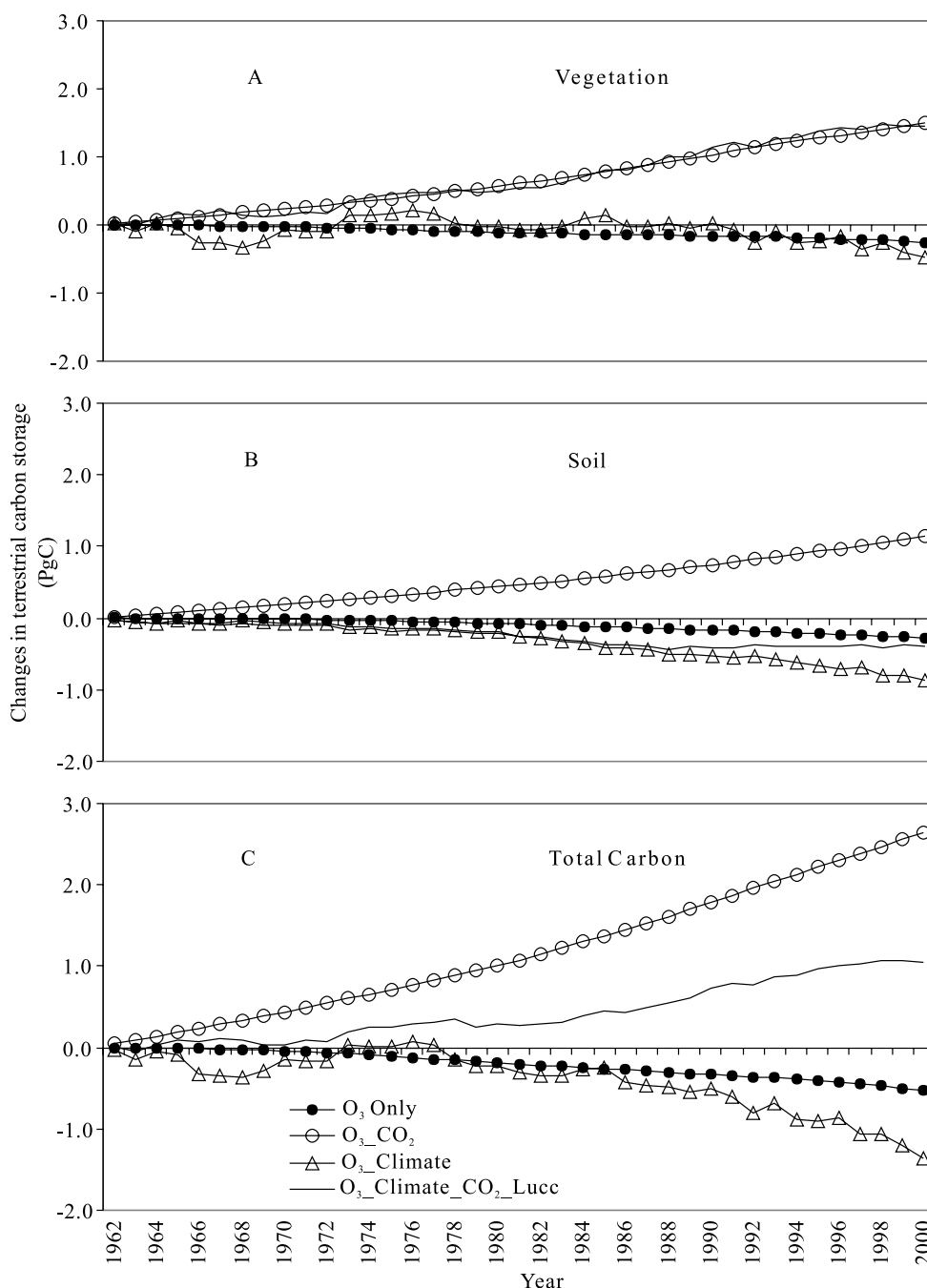
**Figure 7.** Difference in (top) NPP and (bottom) cumulative NCE in 1990s between CLC with O<sub>3</sub> and without O<sub>3</sub> (g m<sup>-2</sup>).



**Figure 8.** Change rate of average annual NPP from 1960s to 1990s under different influencing factors related to O<sub>3</sub> (%). (a) O<sub>3</sub> only, (b) O<sub>3</sub>\_Climate, (c) O<sub>3</sub>\_LUCC, (d) O<sub>3</sub>\_CO<sub>2</sub>, (e) O<sub>3</sub>\_Climate\_LUCC\_CO<sub>2</sub>, and (f) Climate\_LUCC\_CO<sub>2</sub>.



**Figure 9.** Changes in annual NPP during 1961–2000 as forced by different factors and their combinations.



**Figure 10.** Annual changes in terrestrial carbon storage in China from 1961 to 2000 under four influencing factors.

Furthermore, our work shows different sensitivities for different PFTs, in part because we incorporated the *Reich* [1987] dose-response functions into DLEM. Deciduous forests and dry farmland crops are relatively more sensitive to ozone than other PFTs. Dry farmland showed more reduction in yield than paddy farmland. It indicates an important need to further study variations among PFTs and the underlying mechanisms.

**4.2. Ozone and Its Interactive Effects on Net Primary Productivity and Carbon Storage**

[28] In our study, average annual NPP decreased 0.01 PgC/a and accumulated NCE was  $-0.92$  PgC (Figure 9 and Table 4)

from 1961 to 2000 with the effect of  $O_3$  only. Similar to most field experiments in US, Europe and China and other model results [e.g., *Heagle*, 1989, and references therein], our results show that ozone has negative effects on terrestrial ecosystem production due to direct ozone-induced reductions in photosynthesis.

[29] The combined effects of  $O_3$ ,  $CO_2$ , climate, and land use show very different results.  $O_3$  may compensate for  $CO_2$  fertilization and result in NPP losses in different plant types over time across China (Figures 8–10). Climate variability increased the ozone-induced reduction of carbon storage and led to substantial year-to-year variations in carbon

fluxes (NPP and NCE). These results are consistent with many previous studies [e.g., *Cao and Woodward*, 1998; *McGuire et al.*, 2001; *Tian et al.*, 1998b, 1999, 2003]. There is a direct positive effect of elevated CO<sub>2</sub> on photosynthesis and biomass production [e.g., *Agrawal and Deepak*, 2003], as well as reduced stomatal conductance. Climate warming increased decomposition, resulting in continuous loss of soil and total carbon storage (Figures 10b and 10c) since 1990, although annual precipitation was substantial during this period (Figures 6c and 6d). China is influenced by a monsoon climate so that summer monsoons bring most of the annual precipitation [*Fu and Wen*, 1999]. The combined effects of changes in ozone concentration and climate warming in arid areas may result in larger variability in productivity.

[30] Similarly, the contribution of land use change to the terrestrial carbon budget varied over time and among different ecosystem types [e.g., *Houghton and Hackler*, 2003]. In our study, we took into account the combined effects of land use change with historical ozone concentration, atmospheric CO<sub>2</sub> concentration, and climate variability. When considering the effects of land use with ozone, three aspects need to be addressed. First, transformations of different land use types, such as the conversion from forest to crop and regrowth of natural vegetation after cropland abandonment, could result in carbon loss or carbon uptake [e.g., *Tian et al.*, 2003]. The other two are the sensitivities of different biomes to ozone exposure and agriculture management. The former results in different carbon loss rates; however, the latter's effects combined with ozone pollution on carbon storage are related to changing soil environment such as water and nitrogen conditions. In addition, dry farmland and C<sub>3</sub> grass are more sensitive than paddy farmland and C<sub>4</sub> plant types, which could better explain the field study results of the relationship between ozone effects and photosynthesis and stomatal conductance. *Reich's* [1987] study indicated that a secondary response to ozone is possibly a reduction in stomatal conductance, as the stomata close in response to increased internal CO<sub>2</sub>. *Tjoelker et al.* [1995] found a decoupling of photosynthesis from stomatal conductance as a result of long-term exposure to ozone. Such a decoupling implies that ozone-induced reductions in photosynthesis would also be accompanied by decreased water use efficiency (WUE), resulting in even larger reductions in productivity, particularly at arid sites, although many studies indicate that drought-induced stress could reduce ozone stress [*Smith et al.*, 2003]. Unlike C<sub>4</sub> photosynthesis, which adds a set of carbon-fixation reactions that enable some plants to increase photosynthetic water use efficiency in dry environments, C<sub>3</sub> grass and dry farmland in China are always water limited and are more sensitive to ozone exposure. In addition, the modeling studies of *Felzer et al.* [2004] indicated that ozone pollution can reduce more NPP with fertilizer application. In contrast to land use change in the eastern U.S. [*Felzer et al.*, 2004], it is necessary to consider how to manage irrigation in arid areas because there are a lot of moisture-limited regions that require irrigation in China. Reasonable irrigation management can both enhance the water use efficiency and modify the ozone damage.

[31] Besides the environmental factors discussed above, recent reviews of the global carbon budget also indicate that terrestrial ecosystem productivity could be affected by other

changes in atmospheric chemistry, such as nitrogen deposition and aerosols [e.g., *Pitcairn et al.*, 1998; *Bergin et al.*, 2001; *Lü and Tian*, 2007]. These changes can directly change carbon storage. For example, aerosols and regional haze may also reduce ecosystem productivity by decreasing solar radiation and changing climate conditions [e.g., *Huang et al.*, 2006]. Nitrogen deposition could also affect terrestrial carbon storage in a complex way [*Aber et al.*, 1993]. For example, many terrestrial ecosystems in middle and high latitudes are nitrogen limited [e.g., *Melillo*, 1995], and the increasing effect of elevated CO<sub>2</sub> on photosynthesis could be decoupled by limited nitrogen concentration. However, increasing nitrogen deposition could reduce total plant phosphorus uptake [e.g., *Cleland*, 2005] and nitrogen deposition can bias the estimates of carbon flux and carbon storage either high or low depending on the nature of the interannual climate variations [*Tian et al.*, 1999]. In this study, we ran the model with a closed nitrogen cycle because a database containing a time series of nitrogen deposition was not available for our transient analyses. To completely understand the effects of air pollution conditions on terrestrial ecosystem productivity, future work should take these atmospheric chemistry factors into account.

### 4.3. Uncertainty and Future Work

[32] An integrated assessment of O<sub>3</sub> impacts on terrestrial ecosystem production at a large scale basically requires the following types of information: (1) O<sub>3</sub> data set that reflects the air quality in the study area and other environmental data such as climate (temperature, precipitation and radiation), plant (types, distribution and parameters) and soil (texture, moisture, etc.) information; (2) mechanisms of O<sub>3</sub> impacts on ecosystem processes, which describe relationships between air-pollutant-dose and ecophysiological processes such as photosynthesis, respiration, allocation, toleration, and competition; and (3) an integrated process-based model, which is able to quantify the damage of ozone on ecosystem processes. To reduce uncertainty in our current work, future work needs to address the following: First, we used one set of O<sub>3</sub> data from a combination of global atmospheric chemistry models, which have not been validated well against field observations because of limited field data. We found that the seasonal pattern of our simulated ozone data (AOT40) was the same as the limited observation data sets [*Wang et al.*, 2007], with the highest ozone concentrations in summer. However, we still need observed AOT40 values to calibrate and validate our O<sub>3</sub> data set in the future. Second, the ozone module in our DLEM focuses on the direct effects on photosynthesis and indirect effects on other processes, such as stomatal conductance, carbon allocation, and plant growth. The quantitative relationship between O<sub>3</sub> and these processes remains untested by field studies. Third, in our simulations, we simulate land use change accompanying optimum fertilization and irrigation management. It is hard to separate the contributions of land use change from their combination with fertilizer and irrigation. Especially, according to the studies of *Felzer et al.* [2004, 2005], the ozone effect with fertilizer management could increase the damage to ecosystem production. However, irrigation management in dry lands could reduce the negative effect of ozone [*Ollinger et al.*, 1997]. In our model, crops were classified as dry

farmland and paddy farmland which improved the crops' simulation compared to previous process-based models, but different crop types, such as spring wheat, winter wheat, corn, rice, and soybean, have large differences in sensitivity to ozone and could result in different ecosystem production changes due to the effects of ozone [e.g., Heck, 1989; Wang *et al.*, 2007]. It is needed to improve the agricultural ecosystem module by including different crop types and varied managements (fertilizer, irrigation, tillage, and so on) to study the ozone effect.

[33] To accurately assess impacts of ozone and other pollutants on NPP and carbon storage on a regional scale, it is needed to improve both observation data in China and ecosystem model. So the future research may focus on the following: (1) validation of simulation results with site data; (2) refinement of present ozone data with field observations and comparison of present ozone data with other simulated ozone data; (3) development of the ozone module by coupling the effects of ozone on LAI and stomatal conductance; (4) improvement of the process-based agricultural ecosystem model to study the effects of air pollutants on different crop types; and (5) inclusion of other air pollutants (e.g., aerosols and nitrogen deposition) in the model.

## 5. Conclusions

[34] This work investigated tropospheric O<sub>3</sub> pollution in China and its influence on NPP and carbon storage across China from 1961 to 2000 by using the Dynamic Land Ecosystem Model (DLEM). Our simulated results show that elevated tropospheric O<sub>3</sub> concentration has led to a mean 4.5% reduction in NPP nationwide during the period of 1961–2000. Our simulations suggest that the interactions of ozone with increasing CO<sub>2</sub>, climate change, land use and management are significant, and that the interaction of ozone with the climate and land use change can cause terrestrial ecosystems to release carbon to the atmosphere. Ozone effects on NPP varied among plant functional types, ranging from 0.2% to 6.9% during 1961 to 2000. Dry farmland, C<sub>3</sub> grasses, and deciduous forests are more sensitive to ozone exposure than paddy farmland, C<sub>4</sub> grasses, and evergreen forests. In addition, following ozone exposure experiments, we allowed crops to be more sensitive than deciduous trees and deciduous trees to be more sensitive than coniferous trees, and therefore had the highest mean reduction (over 14.5%) since the late 1980s. Spatial variations in ozone pollution and ozone effects on NPP and carbon storage indicate that eastern-central China is the most sensitive area. Significant reduction in NPP occurred in northeast, central, and southeast China where most crops are planted. Direct and indirect effects of air pollutants on ecosystem production, especially on agriculture ecosystem carbon cycling, should be considered in the future work in China. Lack of ozone observation data sets and sensitivity experiments of different PFT<sub>s</sub> could result in uncertainties in this study. To accurately assess the impact of elevated O<sub>3</sub> level on NPP and carbon storage, it is necessary to develop an observation network across China to measure tropospheric O<sub>3</sub> concentration and its effects on ecosystem processes, and then to enhance the capacity of process-based ecosystem models by rigorous field data-model comparison.

[35] **Acknowledgments.** This research is funded by NASA Interdisciplinary Science Program (NNG04GM39C). We thank Dengsheng Lu, Art Chappelka, Chelsea Nagy and two anonymous reviewers for their constructive suggestions and comments.

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