ECOSYSTEMS

Century-Scale Responses of Ecosystem Carbon Storage and Flux to Multiple Environmental Changes in the Southern United States

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ABSTRACT

Terrestrial ecosystems in the southern United States (SUS) have experienced a complex set of changes in climate, atmospheric CO_2 concentration, tropospheric ozone (O_3), nitrogen (N) deposition, and land-use and land-cover change (LULCC) during the past century. Although each of these factors has received attention for its alterations on ecosystem carbon (C) dynamics, their combined effects and relative contributions are still not well understood. By using the Dynamic Land Ecosystem Model (DLEM) in combination with spatially explicit, long-term historical data series on multiple environmental factors, we examined the century-scale responses of ecosystem C storage and flux to multiple environmental changes in the SUS. The results indicated that multiple environmental changes shifted SUS ecosystems from a C source of 1.20 ± 0.56 Pg (1 Pg = 10^{15} g) during the period 1895 to 1950, to a C sink of 2.00 ± 0.94 Pg during the period 1951 to 2007. Over the entire period spanning 1895–2007, SUS ecosystems were a net C sink of 0.80 ± 0.38 Pg. The C sink was primarily due to an increase in the vegetation C pool, whereas the soil C pool decreased during the study period. The spatiotemporal changes of C storage were caused by changes in multiple environmental factors. Among the five factors examined (climate, LULCC, N deposition, atmospheric CO_2, and tropospheric O_3), elevated atmospheric CO_2 concentration was the largest contributor to C sequestration, followed by N deposition. LULCC, climate, and tropospheric O_3 concentration contributed to C losses during the study period. The SUS ecosystem C sink was largely the result of interactive effects among multiple environmental factors, particularly atmospheric N input and atmospheric CO_2.

Key words: climate change; carbon storage and flux; land use change; Dynamic Land Ecosystem Model (DLEM); southern United States.

INTRODUCTION

The southern United States (SUS) is defined as the 13 states from the Mid-Atlantic coast west to Texas and Oklahoma, representing approximately 24% of the total land area, 25% of agricultural land, and 60% of forest land in the continental US (Wear 2002; Chen and others 2006). This region has experienced complex changes in multiple environmental factors, such as climate change, air pollution, and land-use and land-cover change (LULCC) during the past 300 years (Houghton and others 1999; Wear 2002; Karl and others 2009; Tian and others 2010a). Although recent studies suggest that this region is the largest carbon (C) sink among the six major bioclimatic regions in the US and has the greatest potential to be a significant C sink in the future (Schimel and others 2000; Birdsey and others 2006; Malmsheimer and others 2008), mechanisms underlying estimates of the long-term C balance of this region are still not well understood.

Changes in climate have been linked to both increases and decreases in plant growth and C storage in terrestrial ecosystems. For instance, large-scale droughts have reduced regional net primary productivity and C storage (Zhao and Running 2010; Chen and others 2012). Continued regional warming may result in C loss from ecosystems through enhanced plant and soil respiration (Woodwell and others 1995; Chen and Tian 2005), but increased nitrogen (N) availability has also been associated with increased plant growth (Tian and others 1999; Melillo and others 2002) and a lengthening of the active growing season (Myneni and others 1997). Elevated atmospheric CO_2 and N deposition have been shown to stimulate photosynthesis in a variety of plant species and ecosystems (Curtis and Wang 1998; Ellsworth 1999; Norby and others 2005; Luo and others 2008). However, it is not clear how long the CO_2 fertilization effects will last in cases where N or other nutrients become limiting (Oren and others 2001; Luo and others 2004; Johnson 2006). LULCC can influence ecosystem C dynamics in several ways including the direct impacts of biomass removal, for example, burning (Houghton and others 1999; Casperson and others 2000); changes in land management practices (for example, fertilization or irrigation) (Ollinger and others 2002); and indirect legacies such as litter decay, disruption of nutrient and water cycles, and wood product decay on the ecosystem structure (for example, species composition and biodiversity) and function (for example, plant regrowth, N and water cycles) as reported by Ramankutty and others (2007). The impacts of tropospheric ozone (O_3) concentration on regional C dynamics have received less attention but current studies indicate that elevated O_3 concentration has significantly reduced C sequestration and net primary productivity (NPP) of terrestrial plants, particularly agricultural crops, which could offset the positive effects of elevated atmospheric CO_2 and N deposition (Chameides and others 1994; Chapelka and Samulson 1998; Ollinger and others 2002; Felzer and others 2004; Ren and others 2007a, 2011a, b).

Although site-level research has provided valuable insights into the ecological impacts of individual environmental factors, the magnitude and spatial patterns of these factors and their interactions with regional C dynamics are not well understood in the SUS. Simple extrapolations based on contemporary datasets (for example, Houghton and others 1999; Han and others 2007) may not be robust since the spatial–temporal heterogeneity of environmental constraints, interactions among multiple factors, and long-term legacy effects are ignored. Most previous studies in this region have focused only on individual or a small subset of environmental controls over a short period (for example, Houghton and others 1999; Schimel and others 2000; Chen and others 2006). Both theoretical research and field experiments have revealed that ecosystem responses to environmental changes are not necessarily additive and could have complex, interactive effects on ecosystem structure and functioning (Oren and others 2001; Ollinger and others 2002; Luo and others 2008; Tian and others 2011a, b). Zhou and others (2008) and McNulty and Boggs (2010) have also suggested that ecosystem responses to multifactor environmental changes could be controlled by complex C, N, and water cycling processes.

We applied a process-based ecosystem model (Dynamic Land Ecosystem Model, DLEM), which fully couples C, N, and water cycles (Tian and others 2010a, 2011a, b) to assess the impacts of multiple environmental factors (for example, climate, atmospheric CO_2, tropospheric O_3, N deposition, and LULCC) and their contributions to C fluxes in the SUS. The two objectives of this study are to quantify the magnitude and spatial–temporal patterns of C storage and fluxes, and estimate relative contributions of multiple environmental factors to long-term terrestrial C fluxes in the SUS using long-term, spatially explicit environmental data and model simulations.

METHODS

Study Region

The SUS extends roughly from 75° to 100° west longitude and from 30° to 37° north latitude, including 13 states: Alabama, Arkansas, Florida, Georgia, Kentucky, Louisiana, Mississippi, North Carolina, Oklahoma, South Carolina, Virginia, Tennessee, and Texas (Figure 1). Elevations within the region range from sea level along the Gulf of Mexico and Atlantic coasts to more than 1,800 m in the Appalachian Mountains. Overall, the climate is temperate, becoming largely subtropical near the coast. The dominant forest types in this region are temperate evergreen needleleaf and deciduous broadleaf.

Model Description, Parameterization, and Corroboration

The DLEM is a process-based terrestrial ecosystem model that simulates daily C, N, and water cycles as influenced by changes in atmospheric chemistry (O_3, CO_2, and N deposition), climate, LULCC, and disturbances (for example, fire, hurricane, and harvest). The DLEM couples multiple processes and their interactions through five core components (Figure 2): (1) biophysics, (2) plant physiology, (3) soil biogeochemistry, (4) dynamic vegetation, and (5) disturbance, land-use and management. The biophysical component includes instantaneous exchanges of energy, water, and momentum with the atmosphere. Micrometeorology, canopy physiology, soil physics, radiative transfer, and water and energy flow are represented by the model. The DLEM simulates major physiological processes such as photosynthesis, respiration, and carbohydrate allocation among various parts (root, stem, and leaf), as well as processes such as N uptake and transpiration. Mineralization, nitrification, denitrification, aerobic and anaerobic decomposition are simulated in the soil component so that the DLEM is able to estimate simultaneous emissions of multiple trace gases (CO_2, CH_4, and N_2O). The DLEM has also been used to quantify water, C, and N cycles in managed ecosystems, including cropland, forest plantations, and pasture. Detailed information on how these ecosystem processes are modeled can be found in Tian and others (2010a).

The uptake of atmospheric CO_2 by vegetation during photosynthesis is represented by gross primary production (GPP) in the DLEM. Carbon dioxide is returned to the atmosphere from the autotrophic respiration of plants (R_A) and through heterotrophic respiration (R_H) associated with decomposition. Net primary production (NPP) is calculated as the difference between GPP and R_A. The net C exchange (NCE) of CO_2 between the terrestrial biosphere and the atmosphere from natural ecosystem metabolism is represented by net ecosystem production (NEP), which is calculated as
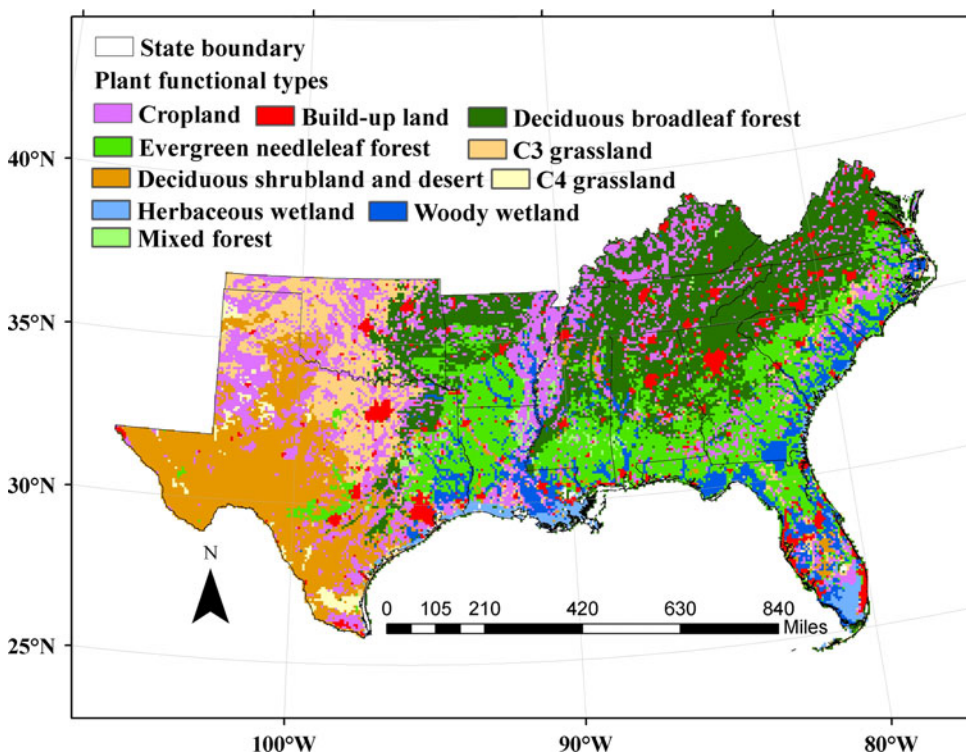

Figure 1. The state boundaries and major plant functional types in the SUS.
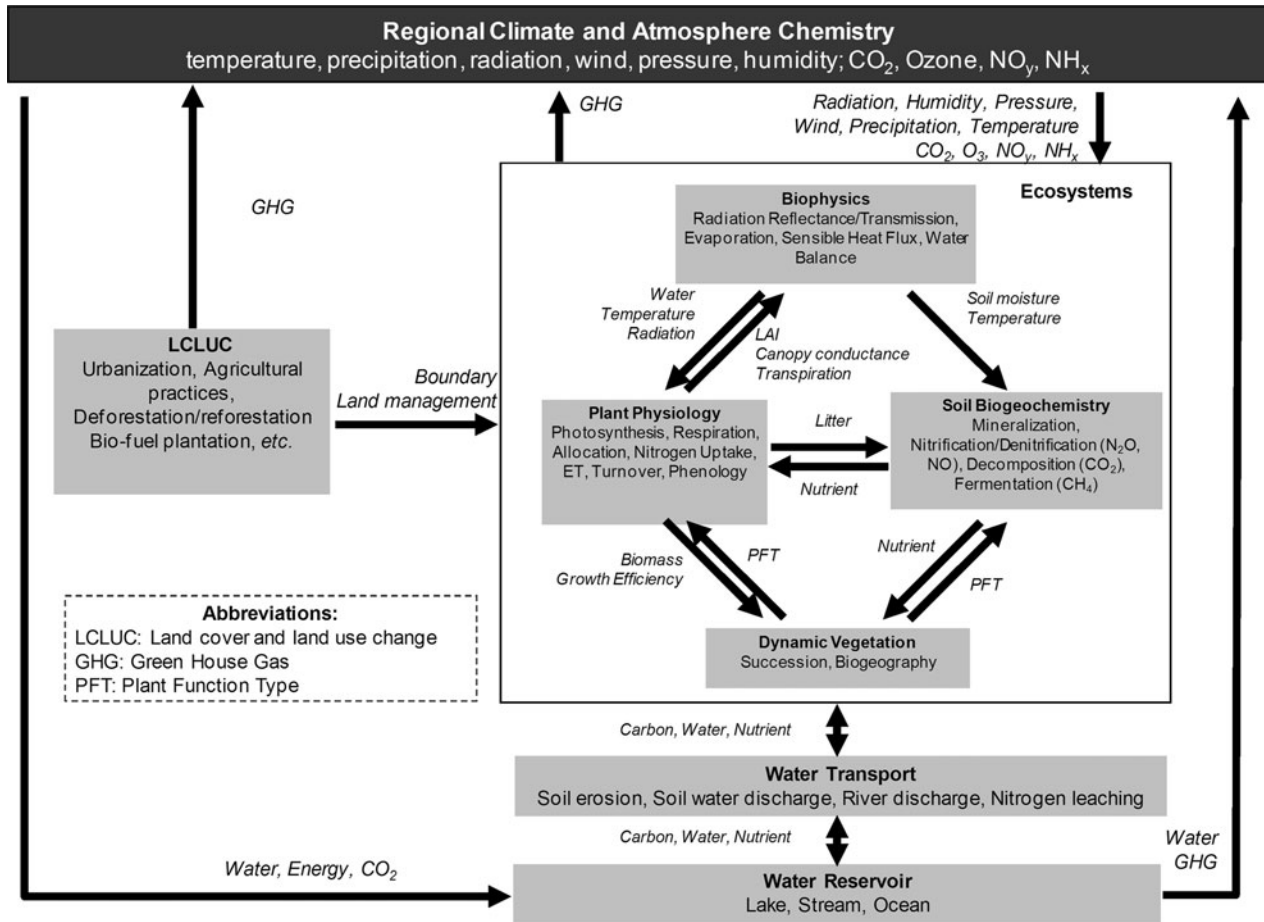
Figure 2. The schematic framework of the DLEM (Dynamic Land Ecosystem Model).

the difference between NPP and R_H. The DLEM also accounts for the C fluxes during land conversion among different plant functional types and the sum of C emissions (E_C) from decay of agricultural and wood products (E_P). In addition, CH_4 emissions (E_{CH_4}) are deducted from the C fluxes. Thus, the NCE is calculated as follows: NCE = GPP − R_A − R_H − E_C − E_P − E_{CH_4} = NEP − E_C − E_P − E_{CH_4}. A positive value of NCE represents a gain of C in terrestrial ecosystems, whereas a negative value represents a loss.

Model parameterization for the SUS has been described in detail in Tian and others (2010a). The DLEM has been corroborated for C and water fluxes (Tian and others 2010a, 2011a, b; Liu and others 2008, 2012; Richardson and others 2012; Sulman and others 2012; Huntzinger and others 2012), N storage and fluxes (Lu and others 2012; Tian and others 2010b), and soil C and N transformation processes and trace greenhouse gas emissions (Tian and others 2010b, 2011a; Xu and others 2010). Zhang and others (2010) also corroborated the simulated forest vegetation, litter, coarse woody debris (CWD), and total (vegetation + soil + litter + CWD) C stocks for each SUS state against corresponding observational data from USDA Forest Service Forest Inventory and Analysis (FIA) collected from 1987 to 1997 (Data source: Birdsey and Lewis 2003). The regression between simulated and observed total C stocks in 1997 shows that DLEM was able to accurately simulate total C stocks across the SUS (Figure 3A; $P < 0.01$; $R^2 = 0.73$; slope = 1.13; coefficient of variation of root mean square deviation, CV(RMSD) = 0.094). In addition, four major vegetation types were selected to corroborate DLEM-simulated daily NEP patterns in this study (Figure 3B–E). Compared to observations, the DLEM-simulated NEP tended to be more biased at extreme high or low values. However, the model generally captured the daily patterns for evergreen needleleaf forest (US-Dk3; $R^2 = 0.30$; CV(RMSD) = 1.59; $n = 2920$), deciduous broadleaf forest (US-Dk2; $R^2 = 0.60$; CV(RMSD) = 1.22; $n = 1825$), C_3 grassland (US-Shd; $R^2 = 0.54$; CV(RMSD) = 1.24;
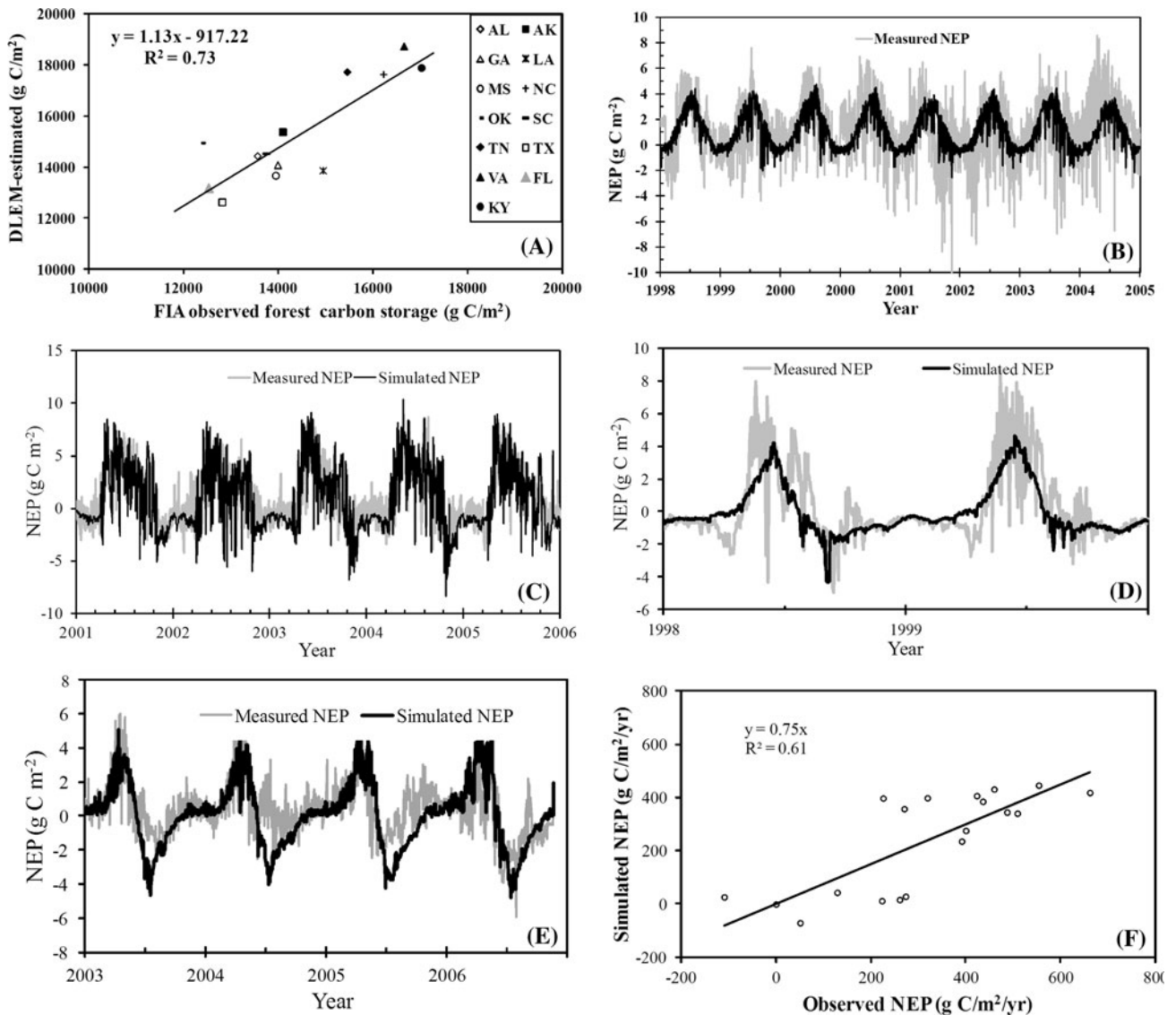
Figure 3. Comparisons of simulated and observed C storage and net ecosystem production (NEP) for different states and land-cover types in the SUS. **A** Correlation between simulated and FIA-observed total ecosystem C storage in different states; **B** Simulated and observed NEP for US-Dk3 AmeriFlux site (Evergreen needleleaf forest); **C** For US-Dk2 AmeriFlux site (Deciduous broadleaf forest); **D** For US-Shd AmeriFlux site (C_3 grassland); **E** For US-ARM AmeriFlux site (Cropland); **F** Correlation between simulated and observed annual C fluxes in the four selected AmeriFlux sites.

$n = 730$), and cropland (US-ARM; $R^2 = 0.44$; CV(RMSD) = 1.39; $n = 1419$). At an annual scale, we found a higher correlation and less RMSE between simulated and observed NEP ($R^2 = 0.61$; slope = 0.75; CV(RMSD) = 0.47) for all these sites (Figure 3F), indicating that the model was better able to capture interannual rather than daily variation.

Input Data Description

The model input data included historical LULCC, climate, atmospheric CO_2, tropospheric O_3 con-

centrations, and N deposition (Table 1). The geospatial data were converted to the same spatial scale (8 km × 8 km). A detailed description of sources and the development of input datasets can be found in Chen and others (2006); Zhang and others (2012), and Tian and others (2010a).

Air temperature exhibited an increasing trend from 1895 to the mid-1950s (0.12°C per decade) and then showed a sudden decrease over several years (1955–1960) (Figure 4B). After the 1960s, air temperature increased again with time (0.12°C per decade). Therefore, during the entire study

**Table 1.** Data Sources and Methods used to Generate the Input Data Sets for Model Simulation in this Study

Input data	Unit	Temporal resolution	Methods and data sources
Potential vegetation map	8 categories	Non-transient base map	Aggregated from the NLCD2001 land cover dataset (Homer and others 2007).
Soil texture (clay, silt and sand)	%		Based on the 1 km resolution digital general soil association map (STATSGO) that was developed by USDA Natural Resources Conservation (NRC), while soil texture was derived from the USDA soil texture triangle (Miller and White 1998)
Soil depth	m		
Soil acidity	pH		
Soil bulk density	g/cm^3		
Elevation map	m		Generated from the 7.5 minute USGS National Elevation Dataset (NED). Data available online: http://edcnts12.cr.usgs.gov/ned/ned.html
Aspect map	°		
Slope map	°		
Irrigation map	1/0		From an Irrigated Area Map of the World developed by Thenkabail and others (2006)
Precipitation	mm	Daily	Developed by integrating the daily climate pattern from the North American Regional Reanalysis dataset (Mesinger and others (2006); www.emc.ncep. noaa.gov), and the monthly climate dataset developed by the Parameter-elevation Regressions on Independent Slopes Model (PRISM) (http://prism.oregonstate.edu/)
Maximum temperature	°C		
Minimum temperature	°C		
Average temperature	°C		
Ozone index AOT40[1]	ppb h		Dataset developed by Felzer and others (2004)
CO$_2$	ppmv	Annual	IPCC CO$_2$ concentration data (Enting and others 1994
Nitrogen deposition[2] (NH$_x$)	g N/m^2/y		Developed based on the global N deposition datasets of three periods (1860, 1993, and 2050) generated by Dentener (2006). Annual variations were controlled by the global historical N emission dataset developed by Van Aardenne and others (2001)
Nitrogen deposition (NO$_y$)	g N/m^2/y		
Cropland fertilization	g N/m^2/y		Based on the county-level fertilizer consumption records (Alexander and Smith 1990; Ruddy and others 2006
Cropland conversion	0/1		Value 1 means urban or cropland. Developed with approaches described by Chen and others (2006)
Urbanization	0/1		

[1]AOT40 (ppb h) is the accumulated dose over a threshold of 40 ppb during daylight hours.
[2]N deposition include NH$_x$ (NH$_3$ and NH$_4^+$), and NO$_y$ (all oxidized forms of N other than N$_2$O).

period, there was no obvious trend in air temperature across the SUS. Annual precipitation ranged from 797 mm in 1954 to 1,316 mm in 1973 with huge interannual variability (Figure 4A). The study region experienced significant droughts during the mid-1950s (from 1952 to 1956) and again from 2005 to 2007. Annual mean precipitation in the SUS decreased along an east–west gradient with a semiarid climate in the west and a humid climate in the east. Although precipitation showed a general increasing trend for the entire SUS, precipitation decreased in parts of the eastern portion of the SUS from 1895 to 2007 (Tian and others 2010a). The highest precipitation (>2,000 mm/y) was found in the Great Smoky Mountains region and the lowest in the northwestern Texas (<200 mm/y).

The rate of N deposition, N fertilizer use in croplands, and tropospheric O$_3$ concentrations (AOT40) increased dramatically from the 1950s to the 1970s (Figure 4C, D, F). The increasing trend of N deposition slowed down after 1980 but continued to increase (Figure 4C). Fertilizer N use for cropland reached its peak in 1994 and leveled off thereafter (Figure 4D). After increasing initially, tropospheric O$_3$ remained relatively stable after
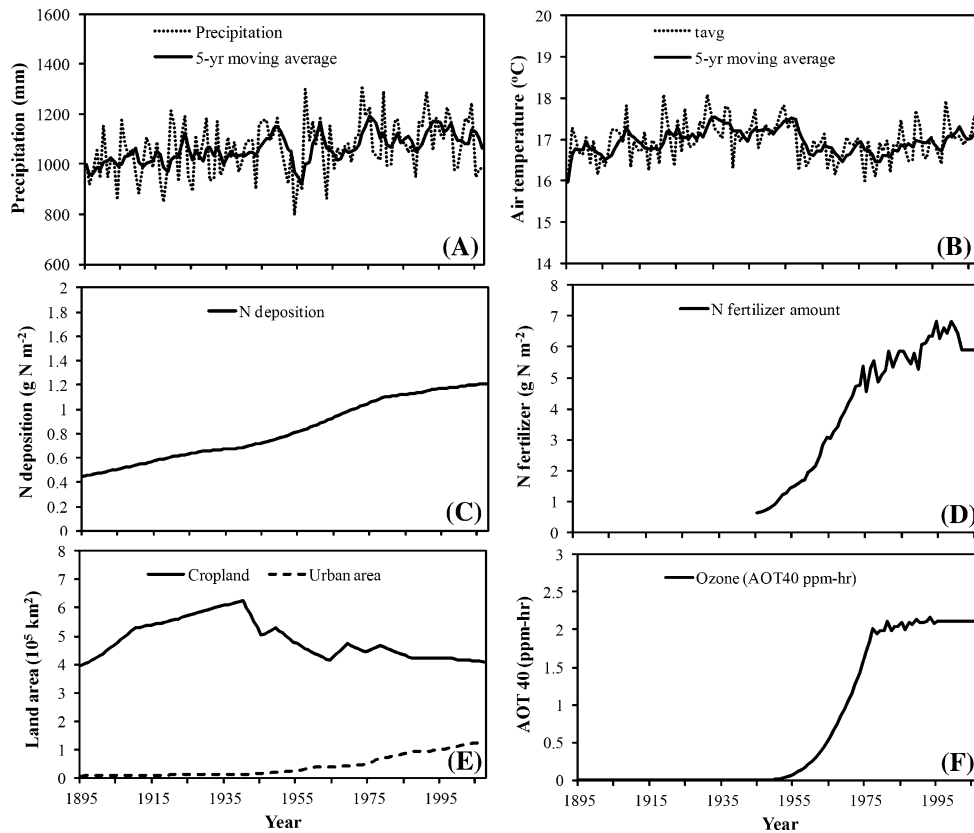
Figure 4. Annual variations of environmental factors in the SUS from 1895 to 2007. **A** Mean annual precipitation (mm); **B** Mean annual temperature (°C); **C** N deposition rate (g N/m^2); **D** N fertilizer amounts in the cropland (g N/m^2); **E** Interannual changes in cropland and urban area from 1895 to 2007 (10^5 km^2); **F** mean exposure to tropospheric ozone stress as measured by AOT40 index (AOT40: hourly accumulated daytime dose over 40 ppm during a month, ppm h).

1980 (Figure 4F). The eastern portion of the study region (that is, Virginia, Kentucky, Tennessee, and North Carolina) received much higher rates of N deposition and O₃ concentrations than the other areas in the study region (Figure 5C–E). The total N deposition rate in Kentucky and the eastern Virginia was as high as 2.0 g N/m^2/y.

The temporal pattern of LULCC was quite complex during the study period (Figure 4E). Before the 1940s, the LULCC was dominated by land conversion from natural vegetation to cropland. Cropland area expanded from 39 million ha to 62 million ha from 1895 to 1940, resulting in decreases in forest (7%), shrubland (13%), and grassland (32%) area over this time period. After 1940, LULCC was characterized as land conversion from cropland to natural vegetation due to cropland abandonment and afforestation/reforestation. The area of forest increased by about 13% from 1940 to the end of 1960s. After the 1960s, cropland area decreased by 13% due primarily to urban area expansion, which doubled from 1980 to 2007. Cropland abandonment primarily resulted in reforestation or afforestation in the southeast portion of the region (for example, Virginia, Georgia) where forests are the dominant native ecosystem type, and in conversion to grassland or shrubland

in the southcentral areas of the SUS (for example, Texas, Oklahoma) (Figure 5G).

Model Simulation and Experimental Designs

The model simulation encompassed the entire SUS from 1895 to 2007 with a spatial resolution of 8 km × 8 km. DLEM was first run to an equilibrium state using mean climate datasets to develop the simulation baseline for C, N, and water pools. Then a 90-year spin-up simulation was conducted using the detrended climate data to eliminate noise caused by simulation shift from the equilibrium to transient mode. The data-detrending approach subtracts the best-fit line from transient climate dataset, and only retains the fluctuations about the trend. Such a dataset is required for stabilizing the simulation before entering the transient mode. Finally, the DLEM simulated daily C fluxes and pool sizes at 8 km × 8 km spatial resolution using the daily transient datasets from 1895 to 2007.

In this study, 16 model simulations were designed to investigate impacts of individual environmental stresses and their interactions on the ecosystem C dynamics in the SUS. There were three types of simulations. The first type was a
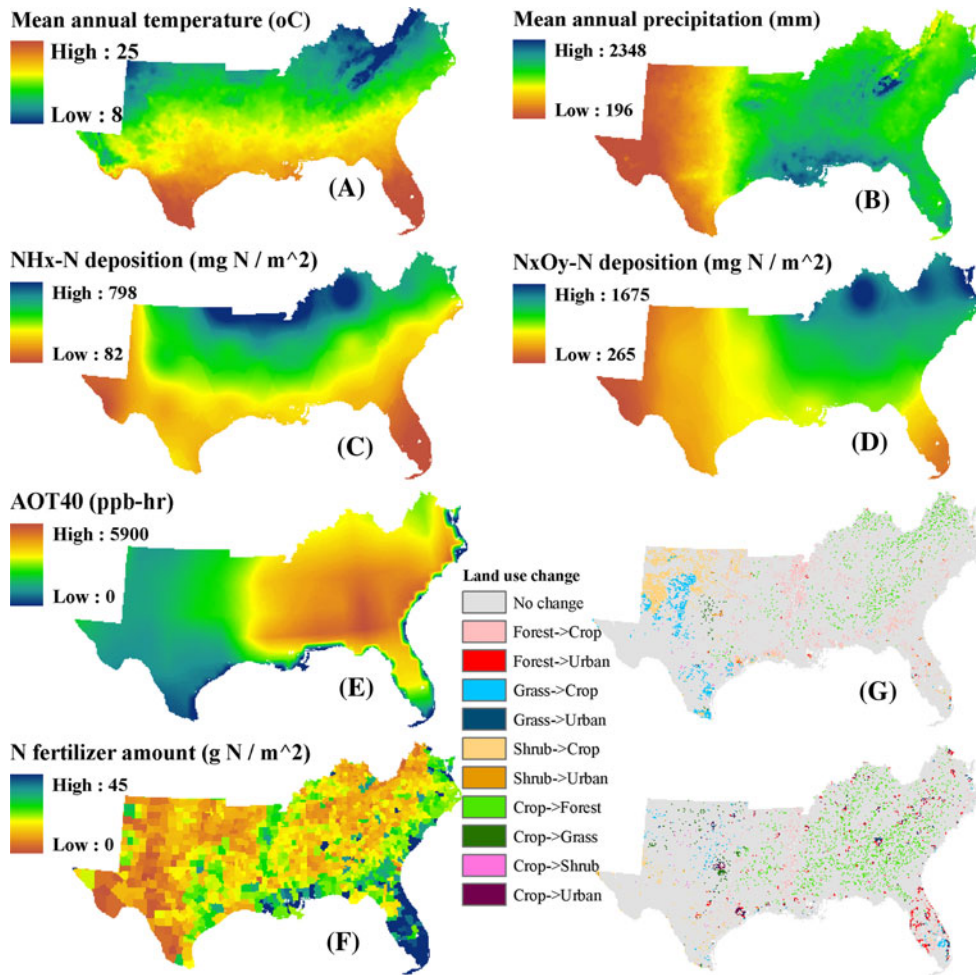
Figure 5. Spatial patterns of multiple environmental change factors in the SUS. (**A**, **B**) daily temperature and annual precipitation from 1895 to 2007, respectively; **C**, **D** annual N deposition rates in 2007 (mg N/m^2. NH$_x$ includes NH$_3$ and NH$_4^+$; N$_x$O$_y$ includes oxidized forms of N other than N$_2$O); **E** mean exposure to tropospheric ozone stress as measured by AOT40 index (ppb h) in 1990s; **F** county-level N fertilizer amounts in cropland in 2000 (g N/m^2); **G** LULCC from 1895 to 1951 and from 1951 to 2007.

single factor simulation (five separate simulations) used to reveal effects of individual environmental factors on the C dynamics. The simulations were (1) CLM: climate change only; (2) CO$_2$: CO$_2$ change only; (3) OZONE: O$_3$ change only; (4) NDEP: N deposition change only; (5) LUC: LULCC only (including land management practices in cropland, such as irrigation and fertilization). The second type was the ALLCOMBINE simulation, which included all five environmental factors to reflect C dynamics in the ''real world.'' The third type was comprised of ten simulations, for example, CO$_2$ + NDEP, CO$_2$ + CLM, CLM + LUC, and so on, developed to estimate the interaction of two individual factors on C fluxes and storage. The total interactive effect among multiple environmental factors was calculated as ALLCOMBINE − CLM − NDEP − CO$_2$ − OZONE – LUC, whereas the two-factor interactive effect was calculated as the combined effects of two factors minus that of each factor individually. For example, the interactive effect between CLM and CO$_2$ was calculated as CLMCO$_2$ − CLM − CO$_2$.

Statistical Analysis

The Spearman's correlation was used to evaluate the consistency between observed and simulated interannual and daily patterns of NCE. Statistical significance ($P < 0.05$) of correlation coefficients and linear regression models was calculated using an F test. CV(RMSD), which is defined as the RMSD normalized to the mean of the observed values, was used as one of the indices for evaluating model performance. In addition, the CV(RMSD) between observed and simulated C storage (for example, vegetation C, soil C, total C) and flux (NCE) was used to estimate uncertainty for simulated regional C storage and flux.

RESULTS AND ANALYSIS

Temporal Changes in C Flux and Storage in the SUS

Simulated results indicated that multiple changes in climate, atmospheric composition, and land use and land cover resulted in a net C sink of

0.80 ± 0.38 Pg (1 Pg = 10¹⁵ g) in the SUS for the entire study period (that is, 1895 to 2007). This sink accounted for 2.7% of the total ecosystem C storage within the SUS in 1895 (Table 2). However, SUS terrestrial ecosystems were a C source of 1.20 ± 0.56 Pg from 1895 to 1950 and a C sink of 2.00 ± 0.94 Pg from 1951 to 2007 (Table 2; Figure 6A).

C in vegetation changed more rapidly than did other C pools (that is, soil, product, and litter) during the study period. Vegetation C decreased by 5.0% from 1895 to 1950 and increased by 14.0% from 1951 to 2007 (Table 2). In comparison, soil organic C decreased by 4.0% for the first period and increased by 3.2% during the second period, respectively. Vegetation C shifted from a C source

Table 2. Changes in the Ecosystem C Storage in the SUS from 1895 to 2007

		Carbon pools				
		VEGC	SOC	LTRC	PRODC	TOTC
Year	1895	8.53	19.40	1.42	0.02	29.37
	1951	8.11	18.62	1.30	0.13	28.17
	2007	9.24	19.23	1.47	0.22	30.17
1895–1951	Difference	−0.42	−0.78	−0.12	0.12	−1.20
	Change rate (%)	−4.96	−4.01	−8.44	658	−4.11
1951–2007	Difference	1.14	0.61	0.17	0.09	2.00
	Change rate (%)	14.05	3.25	13.22	67.71	7.12
1895–2007	Difference	0.72	−0.17	0.05	0.21	0.80
	Change rate (%)	8.38	−0.90	3.66	1170	2.72

VEGC: vegetation carbon; SOC: soil organic carbon; LTRC: litter carbon; PRODC: carbon in the harvested products (for example, crop yields, wood products); TOTC: the total carbon storage (including 0.21 Pg C emitted as CH₄).
Unit: Pg C; 1 Pg = 10¹⁵ g.
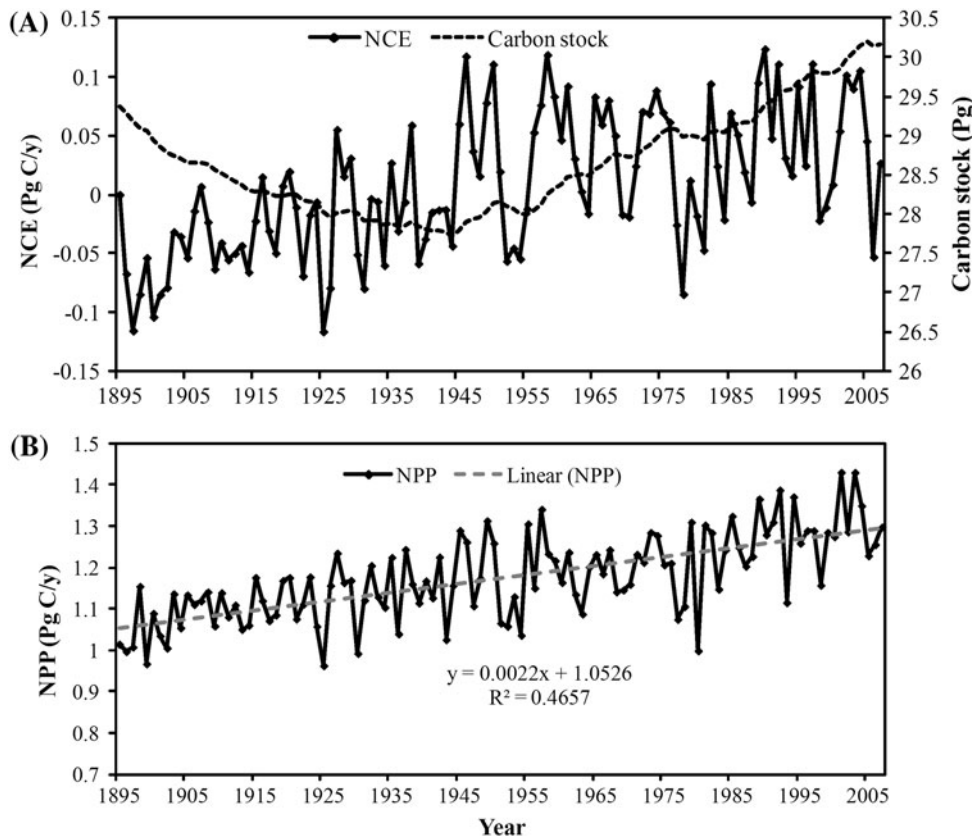
Figure 6. Simulated interannual variation in **A** C storage (including vegetation, soil, litter and product pools, Pg C) and NCE (Pg C/y), and **B** NPP (Pg/y) in the SUS from 1895 to 2007.
1 Pg = 10¹⁵ g.

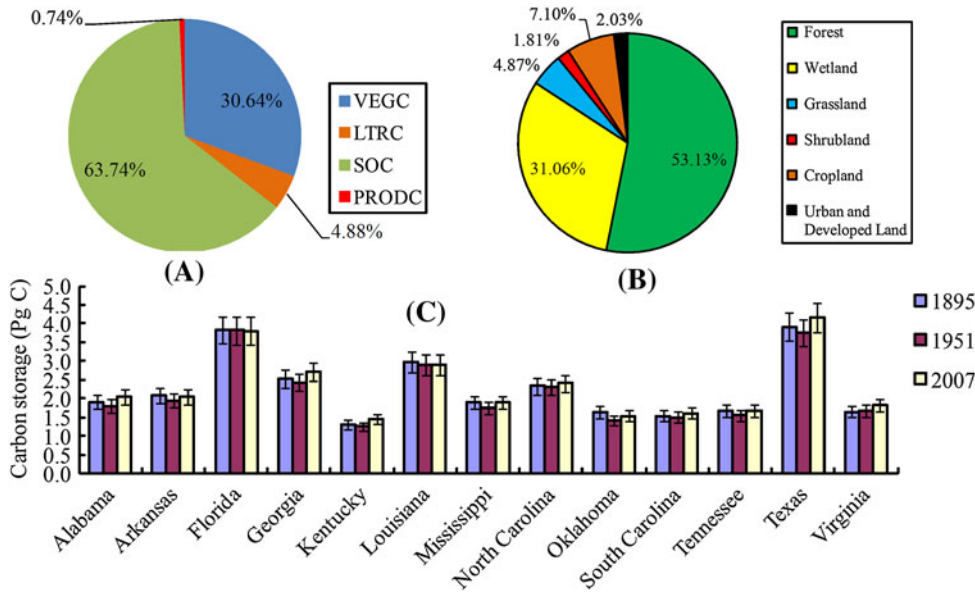In figure B: $y = 0.0022x + 1.0526$, $R^2 = 0.4657$

Figure 7. Distribution of C storage in different **A** C pools in 2007, **B** land-use and land-cover types in 2007, and **C** states (in 1895, 1951, and 2007). VEGC vegetation carbon, LTRC litter carbon, SOC soil organic carbon, PRODC carbon in product pools.
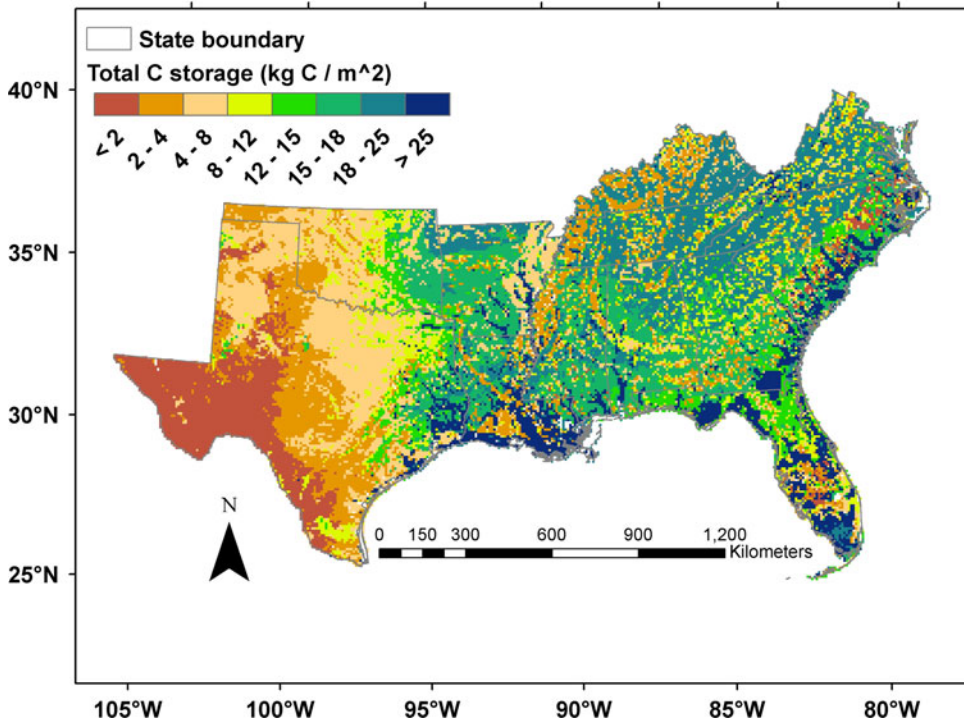

Figure 8. Spatial distribution of C storage (kg C/m^2) in the SUS in 2007 as simulated by the DLEM.

to a C sink in the mid-1940s, whereas soil C shifted to a C sink after the mid-1950s. The vegetation C pool was more responsive to environmental changes (for example, climate, CO_2 concentration) than the soil C pool. Although the vegetation C pool size was only about 48% of the soil C pool size (Figure 7A; Table 2), variations in the vegetation C pool accounted for a greater proportion of long-term change in C storage in the SUS. The net C sink of 0.80 Pg during 1895 to 2007 resulted from combined C in vegetation, litter, and wood product pools (0.97 ± 0.09 Pg) being larger than C losses (0.17 ± 0.02 Pg) from soil pools.

Spatial Variation in C Storage and Fluxes

Land use, land-cover type, and precipitation were the primary determinants of C density, which increased from west to east (Figure 8). The highest C storage, generally greater than 25 kg/m^2, was in
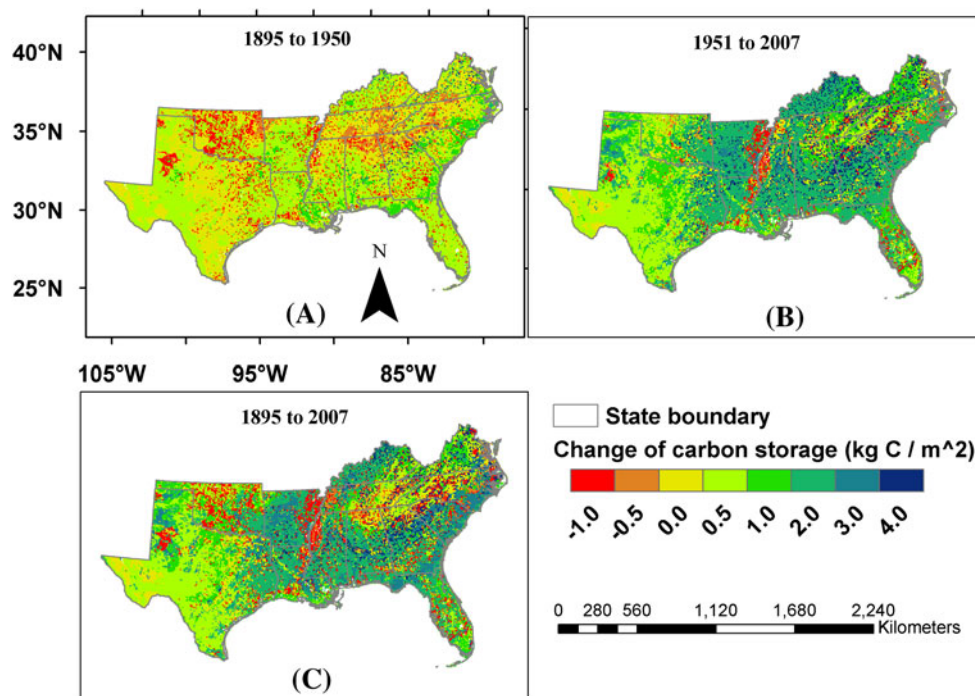
Figure 9. Cumulative NCE from **A** 1895 to 1950, **B** 1951 to 2007, and **C** 1895 to 2007. Negative value means C source, whereas positive value means C sink.

wetland areas. Total C storage was also determined for each state (Figure 7C). During the 2007 regional drought, Texas had the highest terrestrial ecosystem C storage of any state (4.17 ± 0.39 Pg) due to its large land area. This was followed by Florida (3.81 ± 0.36 Pg), which has a large wetland area (about 18% of the total land area) (Figure 1). Kentucky had the lowest C storage (1.46 ± 0.14 Pg).

Both the magnitude and distribution of C sinks and sources varied through time (Figure 9). From 1895 to 1951, the majority of the SUS was either a C source or a weak C sink (Figure 9A). During this period, LULCC was primarily due to conversion from natural vegetation to cropland (Figure 5G), which decreased C storage in most areas including Texas, Oklahoma, Arkansas Alabama and Georgia, and Tennessee (Figure 7C). By contrast, most regions and particularly the southeast area of the SUS region were dominated by strong C sinks between 1951 and 2007 (Figure 9B). For the entire period (that is, 1895 to 2007), C storage increased in most areas of the SUS, with the greatest increases in Kentucky and Virginia (about 10%) and the greatest decrease (6%) in Oklahoma (Figures 7C, 9C). C storage in Tennessee and Florida changed less than 1% during the entire study period (Figure 7C).

Relative Contributions of Environmental Factors to C Storage Changes

The simulation results indicate that changes in the five environmental factors (that is, climate, atmo-

spheric CO_2, tropospheric O_3 concentrations, N deposition, and LULCC) resulted in a net land C sink in the SUS, with substantial interannual variability and spatial variations (Figure 6A). However, it is still unknown how much each factor contributed to this change. Therefore, factorial simulation experiments were used to identify the relative contribution of each factor and their interactions. Among the five environmental factors, LULCC (including N fertilizer application and irrigation in cropland) led to a net C source of 1.26 Pg (Figure 10B, C), accounting for 213% of the net C storage change from 1895 to 2007 (Table 3). Although cropland fertilizer use (Figure 4D) greatly increased C storage (0.45 Pg C), the long-term LULCC legacy effects appear to have largely offset this C sink.

The 30% increase in atmospheric CO_2 concentrations from 1895 to 2007 contributed 1.94 Pg to the SUS C sink. The increases in the atmospheric CO_2 concentrations and N deposition caused the largest contribution of the five factors and consistently increased during the study period (Figure 4C). Nitrogen deposition resulted in a net C sink of 0.48 Pg, contributing about 82% of the net change in C storage (Figure 10B, C; Table 3). Elevated O_3 and climate change were less influential, reducing C storage by 0.58 and 0.40 Pg during the study period. Increased O_3 concentrations primarily influence C fluxes by reducing photosynthetic rates. Under the OZONE experiment, NPP for all the vegetation types was re-
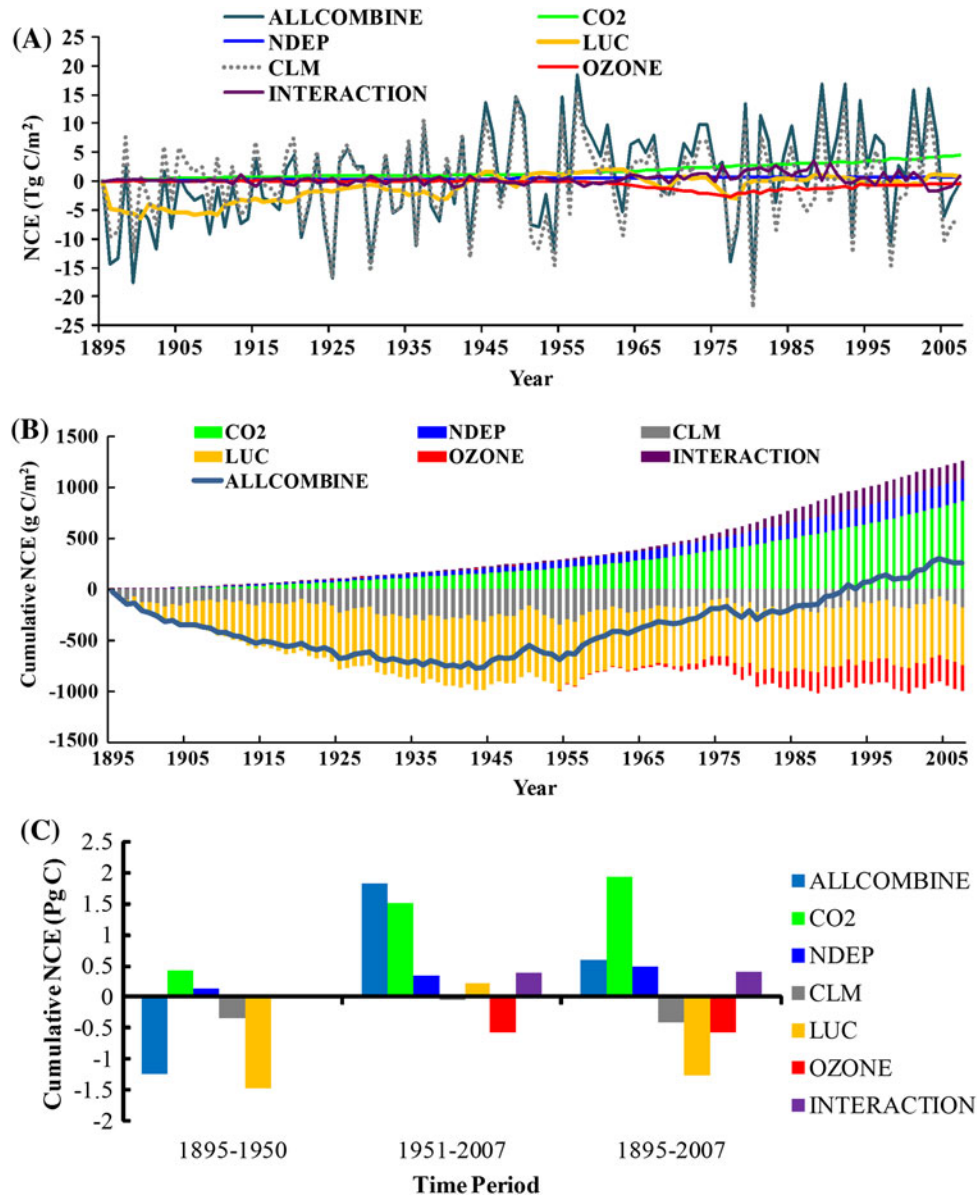
Figure 10. Impacts of environmental factors on annual and cumulative NCE in the SUS from 1895 to 2007 as simulated by the DLEM under different scenarios. **A** The temporal pattern of annual NCE; **B** The cumulative NCE under impacts of different environmental factors and their interactive effect; **C** The relative contribution of multiple environmental factors to NCE during three time periods (1895 to 1950, 1951 to 2007, and 1895 to 2007). INTERACTION means the interactive effect of multiple factors, INTERACTION = ALCOMBINE − OZONE − LUC − CLM − NDEP − CO_2.

duced by 2.5% during the study period. Although the total contribution of long-term climate change to C losses was relatively small, the interannual variation of C fluxes under the combined experiment (ALLCOMBINE) was primarily controlled by climate change (CLM). Correlation analysis indicated that 85% ($R^2 = 0.85$; $P < 0.001$) of C flux variation was explained by climate variability.

Interactive Effects among Multiple Environmental Factors

The interactive effect of all environmental factors resulted in a C sink of 0.40 Pg C over the entire study period (Table 3). Some factors canceled each other out with regard to C sequestration such as the N deposition effect (0.48 Pg C) and climate change effect (−0.40 Pg C). This implied that complex

Table 3. NCE and the Contribution of Different Factors in the SUS from 1895 to 2007

Periods	Storage change (Pg C)	Relative contribution (%)[1]					
		CO_2^3	NDEP	CLM	LUC	OZONE	INTERACTION
1895–1950	−1.24	−33.95	−10.62	27.76	119.18	0.00	−2.29
1951–2007	1.83	82.78	19.22	−2.94	12.04	−31.71	20.58
1895–2007	0.59[2]	328.12	81.93	−67.47	−213.13	−98.35	68.63

[1]*Relative contribution is calculated as the percentage of C storage change induced by each environmental factor to that of the combined factors.*
[2]*C emissions as CH_4 (0.21 Pg C) are excluded in this calculation of C storage change.*
[3]*Simulation experiments: CO_2, NDEP, CLM, LUC, and OZONE were atmospheric CO_2, N deposition, climate, LULCC, and O_3 change only experiments, respectively; INTERACTION: the interactive effects among all factors.*

interactions existed between these factors. The largest positive interactive effect was found between CO_2 and N deposition (0.35 Pg C, Table 4), indicating increasing N deposition could enhance the CO_2 fertilization effect. The largest negative interactive effect was found between climate warming and elevated atmospheric CO_2 (−0.24 Pg C), indicating that warming might weaken the CO_2 fertilization effect on C sequestration. The slight increasing trend in precipitation (Figure 4A) resulted in a relatively large positive interaction (0.20 Pg C) between climate change and N deposition because N deposition rates were related to precipitation. Ozone-climate and O_3–CO_2 interactive effects contributed to C gains in the study region even though their interactions reduced NPP. One cause for the C gains resulting from the CO_2–O_3 interaction was its positive effect on litter quantity, which likely led to increases in soil C storage. No significant interactive effects between other factors were found.

To further elucidate the mechanisms of the interaction between N deposition (N availability) and atmospheric CO_2, we analyzed their effects on natural vegetation types at the biome level, including evergreen needleleaf forest, deciduous broadleaf forest, arid shrubland, and grassland (Figure 11). The results indicate that the CO_2

Table 4. Interactive Effects of Multiple Factors on NCE (Pg C)

Factors	CO_2	NDEP	LUC	CLM	OZONE
CO_2	1.94				
NDEP	0.35	0.48			
LUC	−0.05	0.06	−1.26		
CLM	−0.24	0.20	0.05	−0.40	
OZONE	0.26	−0.07	0.03	0.21	−0.58

Note: The numbers on the diagonal line are the effect of individual factors on NCE during 1895–2007, whereas other numbers are the interactive effect between two individual factors.

fertilization effect on C storage was obviously enhanced by N deposition for all natural vegetation types except grassland (Figure 11E), where N deposition only slightly enhanced the CO_2 fertilization effect. This interactive effect was less responsive at lower N deposition and atmospheric CO_2 levels and increased at higher levels for all biome types except grassland.
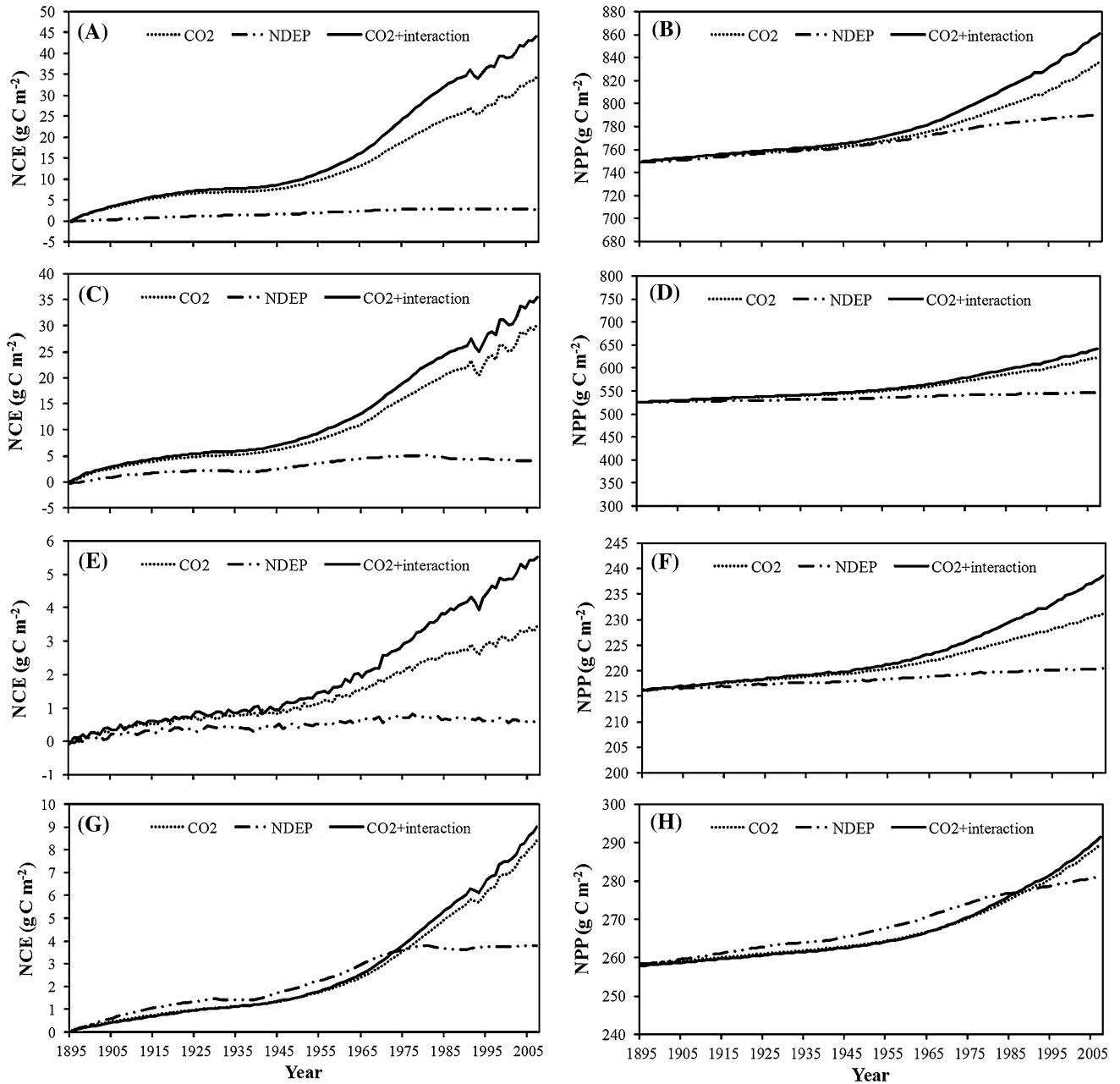
Spatial Variations in C Flux Caused by Changes in Individual Environmental Factors

Although the interannual patterns were relatively simple for NCE as influenced by multiple environmental factors, the spatial patterns were more complex. Atmospheric CO_2 concentration did not vary greatly over space. However, the impacts of elevated CO_2 concentration could result in a C sink ranging from less than 150 g C/m^2 in the west to over 2,500 g C/m^2 in the east during the 1895 to 2007 period (Figure 12A). In general, elevated CO_2 concentrations stimulated more C uptake in forests than other land-cover types. The N deposition rate in the SUS (Figure 5C, D) was generally lower than those in the northeastern US, where N saturation effects induced by excess N deposition have been reported (Pardo and others 2006). N deposition could result in a C sink ranging from 50 to 1,200 g C/m^2 in the SUS (Figure 12B). Cropland was less responsive to increasing N deposition due to the large amount of N fertilizer applied to cropland. Although cropland area in the SUS did not change much from 1895 to 2007, the spatial distribution of cropland changed significantly, with decreased area in the northeast area of the SUS region and increased area in the northwest (Figure 5G). In addition, urban areas increased rapidly since the 1970s. These general LULCC patterns resulted in C sinks in the northeast whereas the northwest was largely a C source (Figure 12C). However, C storage patterns were modified by

Figure 11. Interactive effects between N deposition and atmospheric CO_2 on NCE (g C/m^2) and NPP (g C/m^2) for different biomes in the SUS. **A, B** deciduous broadleaf forest; **C, D** evergreen needleleaf forest; **E, F** shrubland; **G, H** grassland. *Note* CO_2: CO_2 only experiment; NDEP: N deposition only experiment; CO_2 + interaction: CO_2 only effects plus interactive effects between CO_2 and N deposition.

cropland irrigation and N fertilizer use, resulting in C sinks in a few areas in the northwest. Climate change could result in both C sources and sinks in the SUS (Figure 12D). Large C sources found in the northeast area of the SUS region and Florida were primarily due to drought impacts whereas a strong C sink occurred in the west area of the SUS region due to increased precipitation. Increased tropospheric O_3 induced large C sources in the

northeast area of the SUS region (Figure 12E). The influences of tropospheric O_3 concentrations on the photosynthesis of broadleaf forest and cropland increased C emissions in these areas. Interactive effects among multiple environmental factors caused large C sinks in the northeast area of the SUS region (Figure 12F) but resulted in C sources in areas dominated with cropland and wetlands.
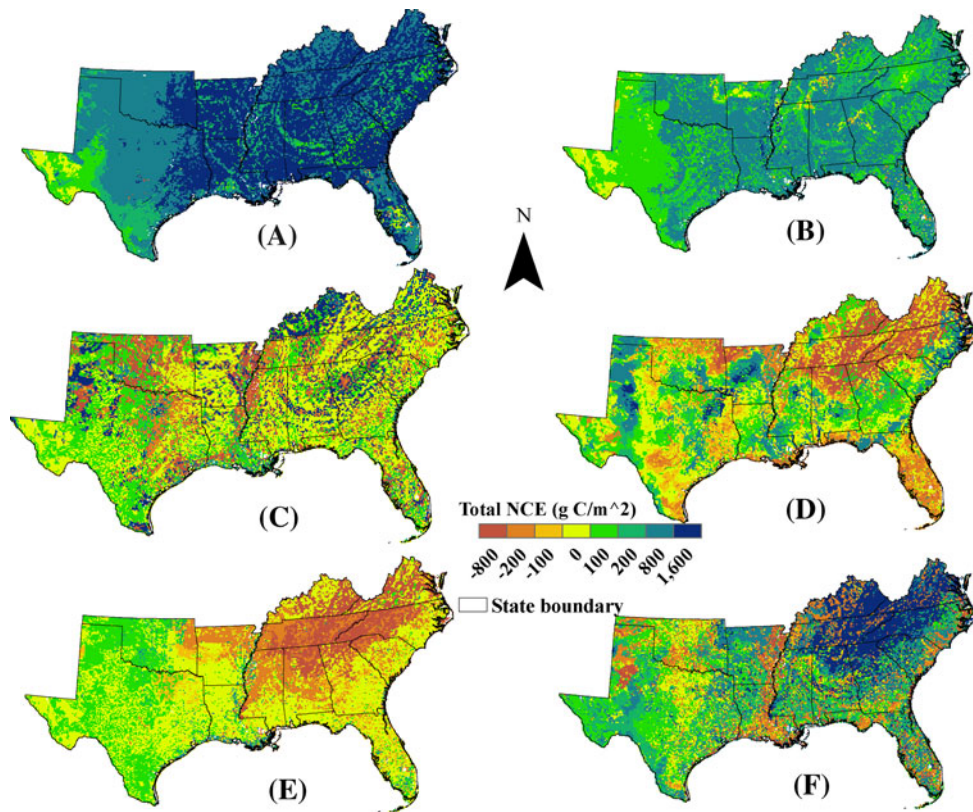
Figure 12. Effects of multiple environmental changes on cumulative NCE (Pg C) in the SUS from 1895 to 2007. **A** CO$_2$ experiment; **B** NDEP; **C** LUC; **D** CLM; **E** OZONE; **F** Interactive effects among multiple factors.

DISCUSSION

Ecosystem C Storage and Flux in the SUS

Han and others (2007) estimated that total C storage was 21 Pg in 11 states of the SUS (Oklahoma and Kentucky were excluded), which is 25% lower than our estimation of 26 Pg C. Despite this difference, both studies reported that Texas has the largest C storage, followed by Florida. The lower value estimated by Han and others (2007) may be partly due to the exclusion of C storage in litter, coarse wood debris, wood products, grassland biomass, and harvested crop products. All these pools have large spatial and interannual variations, and thus could not be simply represented by a regional mean value. Unlike the empirical approach used by Han and others (2007), our study not only directly addressed the spatial and temporal heterogeneity of SUS ecosystems but also the daily variations of C pools in response to environmental changes.

Our simulated NCE for different time periods between 1895 and 2007 was comparable to the reported mean annual NCE from several other studies on national or continental scales (Table 5). We found that the SUS experienced a transition from C source to C sink during the 1950s. In this study, we considered the effects of climate change, atmospheric chemical composition, and LULCC. Most previous work only included a subset of these factors. The study by Schimel and others (2000), for example, did not consider the effect of LULCC, one of the most important factors controlling ecosystem C balance in the SUS since the seventeenth century (Delcourt and Harris 1980). Houghton and others (1999) only focused on the effects of LULCC and ignored the impacts of other environmental factors. Furthermore, our study quantified C fluxes at a higher frequency (daily) and at a higher spatial resolution (8 km × 8 km) compared to the inverse model (for example, Peters and others 2007) or empirical approaches (for example, Pacala and others 2007) used by previous investigators. This approach enabled us to show that the recovery of the soil C pool lagged behind the regrowth of vegetation biomass. For example, the soil in this region has a greater potential to accumulate C than did vegetation. Although total ecosystem C storage has increased by 0.80 Pg C, about 0.20 Pg C has been lost from the soil in the SUS from 1895 to 2007 (Table 3), mainly due to the LULCC legacy effect. This analysis revealed a relatively complex pattern in the dynamics of soil C pools (Table 3).

Table 5. Comparison of Simulated NCE Against the Estimates from Other Previous Studies

Source	Method	Study region	Time period	Mean annual C sink (g C/m²/y)	
				Other study	This study
Ciais and others (1995)	δ^{13}C measurements	Northern mid-latitudes	1992	33–55	50
Houghton and others (1999)	Book Keeping Model	US[1]	1980s	16–38	14
Pacala and others (2007)	Literature review and synthesis	US	Current	53	27[2]
Schimel and others (2000)[3]	Biogeochemical models	Southeast US	1980–1993	15	20
Schimel and others (2001)	Inverse model	North America	1990–1994	32	30
Peters and others (2007)	Inverse model: CarbonTracker	SUS	2000–2007	29.1	23.7

[1]Area of the US is 9.162×10^{12} m.²
[2]The estimate from Pacala and others (2007) includes the C sink due to woody encroachment (120 Tg; 1 Tg = 10^{12}g) and organic sediments accumulated in wetland (23 Tg) and rivers and reservoirs (25 T g) which are not included in our study.
[3]The southeast US defined in this study includes all the states in the SUS except Texas and Oklahoma. Therefore, their estimated C sink is lower than this study.

Relative Contributions of Individual Environmental Change Factors

Some previous studies have suggested that the SUS was a net C sink during the past century. However, most of these studies only included a single or a few factors and could not clearly explain the causes of this C sink. In this study, we included multiple environmental factors to more accurately estimate the spatial and temporal changes in C storage and their causes. Single factor experiments have suggested that fertilization effects from elevated CO_2, N deposition, and N fertilizer consistently stimulated C sequestration, whereas increases in O_3 concentrations led to a consistent C loss since the 1950s. However, the impacts of climate and LULCC could vary through time.

The SUS has experienced three distinct periods in LULCC since the 1700s (Wear 2002; Chen and others 2006; Hansen and others 2010). From 1895 to 1950, the SUS experienced a period of cropland expansion, resulting in a large C emission. We found the LULCC shifted from a C source to a C sink in the early 1950s. This pattern has also been reported by Chen and others (2006) and Houghton and others (1999, 2003). LULCC was a net C sink of 0.23 Pg after 1945, which is about 10% of the reported C sink resulting from LULCC for the continental US (about 2.4 Pg C as estimated by Houghton 2003). From 1895 to 2007, LULCC resulted in a C source of 1.26 Pg, contributing approximately 30% of the C source induced by LULCC for the continental US during approximately the same period (4.1 Pg C as estimated by Houghton 2003). Although cropland area in 1895

was similar to that in 2007 and cropland area abandoned for forests and N fertilizer applications has greatly increased since the 1950s. LULCC still resulted in the largest C source in the SUS from 1895 to 2007. This implied that C storage did not fully recover to the 1895 level from the 1950s to the 2000s. LULCC legacy effects on C sources may be long lasting (for example, 100 years) and therefore short-term studies (for example, Han and others 2008) or static extrapolations (for example, Houghton and others 1999, 2003) could underestimate LULCC impacts.

Our study suggests that elevated CO_2 was the largest contributor to C sequestration in the SUS. However, the impacts of elevated CO_2 may be constrained by other factors such as climate, land-use history, and N availability (Norby and others 2005; Johnson 2006; Luo and others 2004, 2008). Meta-data analysis indicated that a doubling of the CO_2 concentration could stimulate NPP by a median value of 23% for forest (Norby and others 2005) and stimulate C storage within a wide range of environmental conditions. We found that across all vegetation types, a 30% increase in the CO_2 concentration from 1895 to 2007 stimulated NPP by 12% and C storage by 6%, and stimulated forest NPP by 12% and C storage by 12 and 6% in the SUS.

Increased O_3 concentrations decreased NPP by 3% and C storage 2% (0.58 Pg C), for all vegetation types, 3 and 5% for deciduous broadleaf forest, 0.5 and 0.3% for evergreen needleleaf forest, and 7 and 5% for cropland. Our simulated O_3 impacts on NPP and C storage were within the range of field observational data and other model simulations in

the continental US (Heck and others 1984; Pye 1988; Ollinger and others 2002; Felzer and others 2004). In addition, we found that O_3 had the largest impacts on NPP and C storage of deciduous broadleaf forest and cropland, which was also consistent with previous studies (Chameides and others 1994; Chapelka and Samulson 1998; Ollinger and others 2002; Felzer and others 2004; Ren and others 2007b; Zhang and others 2007).

Elevated N deposition resulted in C sinks ranging from 0.44 to 2.0 Pg C/y in global terrestrial ecosystems (Nadelhoffer and others 1999; Churkina and others 2007; Jain and others 2009). Reay and others (2008) also reported that elevated N deposition could sequester 1–2% of the global anthropogenic C emissions (about 8.5 Pg C in 2007, Boden 2009). We found elevated N deposition induced a C sink of 0.48 Pg and increased NPP by 5% for all vegetation types from 1895 to 2007. N deposition contributed 9.6% to the net C change in the SUS, which is within the lower end of the reported range for global terrestrial ecosystems (that is, Nadelhoffer and others 1999), presumably due to a lower N deposition rate in the SUS compared with other regions of the US, and because there were no interacting effects with N deposition in our study. If interacting effects such as elevated CO_2 are included, the total effect would be closer to 0.40 Pg C or about 18% of the net C changes in the SUS in our study. Although N saturation effects (for example, NPP reduction, increased C and N leaching) have been reported in the northeastern US, we did not find this phenomenon in the SUS (Boggs and others 2005), presumably due to the lower N deposition rate and higher rates of N uptake from younger forests in the SUS (Pan and others 2011; Smith and others 2009; Lu and others 2012).

Climate change has been reported to either increase or decrease C storage, depending on approaches used and studied regions (for example, Schimel and others 2000; McGuire and others 2001; Luo and others 2008). In our study, the long-term effect of climate change on C storage was small (−0.40 Pg) compared to the effects of other factors (Table 4; Figure 10B, C). However, we found that interannual climate patterns had an important influence over interannual variations in C fluxes. In addition, climatic patterns varied across the region. Although a general increase in drought intensity and frequency in the SUS has not been reported (Chen and others 2012), drought events still resulted in a C source of 0.16 Pg from 1895 to 2007 and most areas in the northeastern part of the SUS region were C sources in our analysis (Figure 13A). The long-term effect of drought on C
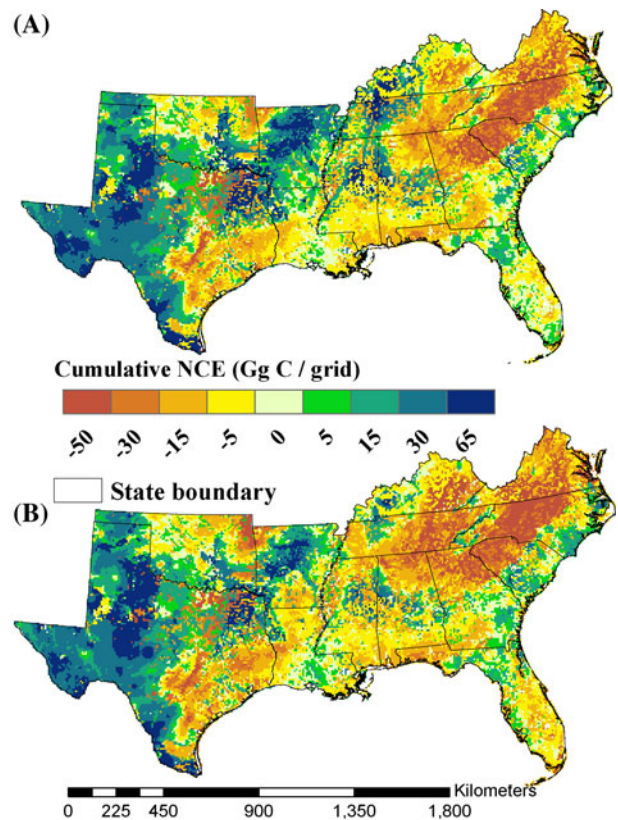

Figure 13. Spatial distribution of accumulated NCE (Gg C/grid; 1 Gg = 10^9 g) as influenced by precipitation only (**A**) and climate change (**B**) from 1895 to 2007.

sources was also enhanced by changes in other climate factors (for example, air temperature and radiation). Climatic extremes, including hot and dry events, could significantly modify the spatial distribution of C sinks and sources in the SUS. For example, our simulation showed that large amounts of C were released during the 1988 to 1989 drought when the SUS experienced a strong La Niňa with a dry winter, whereas an El Niňo in 1991 to 1992 was accompanied by a significant C sink due to the higher precipitation

Interactive Effects among Multiple Environmental Factors

The combined impacts of multiple environmental factors on C dynamics are not simply the additive effects of all the individual factors. Interactive effects have been previously reported (for example, Rastetter and others 1997; Ollinger and others 2002; Shaw and others 2002; McLaughlin and others 2007; Luo and others 2008; Tian and others 2010a, Tian and others 2010b). In our study, the interactions among multiple factors induced a large C sink from 1895 to 2007, which offset C emissions

induced by climate change alone. However, the importance of the interactive effects changed over time. Prior to 1950, the interactive effects were negligible but after 1950 they resulted in a large C sink. Interactions among multiple factors are complex, and more experimental and observational data will be needed to understand them.

The most significant interactive effect occurred between elevated N deposition and atmospheric CO_2, which was also observed by Tian and others (2010b). Our study also found that the interactive effect between N deposition and atmospheric CO_2 increased with elevated N input, implying a stronger N limitation with increasing atmospheric CO_2. This is consistent with the progressive N limitation hypothesis that CO_2 fertilization effects will not be sustained in the long term without external N input (Luo and others 2004; Finzi and others 2006; Johnson 2006).

Uncertainties and Future Research

Although this analysis was intended to simulate the contributions of individual factors and their combined effects, it is also important to recognize the inherent uncertainties in such a regional-scale study. The influences of changes in individual factors on C storage estimates could not be fully corroborated due to lack of adequate long-term observational or experimental data in the study region (Popper 1968; Oreskes and others 1994; Rastetter 1996). The DLEM attempts to include the most important processes controlling regional C dynamics. However, other processes beyond the scope of this model or for which data are lacking may prove to be equally important. For example, forest management (for example, harvest, thinning, and N fertilizer use) and land disturbance (for example, hurricane, fire, and insects) were not considered in this study. These factors alter ecosystem structure and function and influence C fluxes, especially during recent decades. Another uncertainty is the legacy effect of historical disturbances in the study region (Chen and others 2006). In our study, we developed the simulation baseline for all the major C, N, and water pools (for example, vegetation, soil, and litter C and N, soil water content) for 1895 by running the model to equilibrium status, which assumes that the effects of disturbance on vegetation were not significant at that time. This assumption is currently used by many other process-based models (Huntzinger and others 2012) and may lead to overestimation of the baseline C pool size, and therefore the estimated C loss rate due to LULCC in the early twentieth century.

CONCLUSIONS

We explored century-scale regional C dynamics in response to changes in climate, atmospheric CO_2 concentration, tropospheric O_3, N deposition, and LULCC in the SUS from 1895 to 2007 using the DLEM and long-term historical environmental data. Our results suggest that SUS terrestrial ecosystems were a C sink of 0.80 ± 0.38 Pg during this period. An important finding of our study was that SUS ecosystems shifted from a C source between 1895 and 1950 to a C sink from 1951 to 2007. The C sink was primarily due to increased C accumulation in vegetation while the soil C pool declined during the study period. Large spatial variations in the impacts of multiple factors on C dynamics were primarily related to LULCC and precipitation patterns. Elevated atmospheric CO_2 concentration, N deposition, and their interactions were the primary causes of C sinks in our study, whereas changes in tropospheric O_3, LULCC, and climate were the primary causes of C losses. Elevated CO_2 concentration was the largest contributor to C sequestration, whereas LULCC was the largest contributor to C losses. Interactions among multiple factors were complex, but resulted in a large C sink. The interactive effect between N deposition and atmospheric CO_2 continuously increased with elevated N input, implying stronger N limitation with increasing atmospheric CO_2. Additional field experiments and observations are needed to identify the contributions of individual factors and their interactions to C dynamics.

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