Net exchanges of CO₂, CH₄, and N₂O between China's terrestrial ecosystems and the atmosphere and their contributions to global climate warming

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Received 17 April 2010; revised 30 January 2011; accepted 10 February 2011; published 13 May 2011.

[1] China's terrestrial ecosystems have been recognized as an atmospheric CO₂ sink. however, it is uncertain whether this sink can alleviate global warming given the fluxes of CH₄ and N₂O. In this study, we used a process-based ecosystem model driven by multiple environmental factors to examine the net warming potential resulting from net exchanges of CO₂, CH₄, and N₂O between China's terrestrial ecosystems and the atmosphere during 1961-2005. In the past 45 years, China's terrestrial ecosystems were found to sequestrate CO₂ at a rate of 179.3 Tg C yr⁻¹ with a 95% confidence range of $(62.0 \text{ Tg C yr}^{-1}, 264.9 \text{ Tg C yr}^{-1})$ while emitting CH₄ and N₂O at rates of 8.3 Tg C yr⁻¹ with a 95% confidence range of (3.3 Tg C yr⁻¹, 12.4 Tg C yr⁻¹) and 0.6 Tg N yr⁻¹ with a 95% confidence range of (0.2 Tg N yr⁻¹, 1.1 Tg N yr⁻¹), respectively. When translated into global warming potential, it is highly possible that China's terrestrial ecosystems mitigated global climate warming at a rate of 96.9 Tg CO_2 eq yr⁻¹ (1 Tg = 10^{12} g), substantially varying from a source of 766.8 Tg CO_2 eq yr⁻¹ in 1997 to a sink of 705.2 Tg CO₂eq yr⁻¹ in 2002. The southeast and northeast of China slightly contributed to global climate warming; while the northwest, north, and southwest of China imposed cooling effects on the climate system. Paddy land, followed by natural wetland and dry cropland, was the largest contributor to national warming potential; forest, followed by woodland and grassland, played the most significant role in alleviating climate warming. Our simulated results indicate that CH₄ and N₂O emissions offset approximately 84.8% of terrestrial CO₂ sink in China during 1961–2005. This study suggests that the relieving effects of China's terrestrial ecosystems on climate warming through sequestering CO₂ might be gradually offset by increasing N₂O emission, in combination with CH₄ emission.

Citation: Tian, H., X. Xu, C. Lu, M. Liu, W. Ren, G. Chen, J. Melillo, and J. Liu (2011), Net exchanges of CO₂, CH₄, and N₂O between China's terrestrial ecosystems and the atmosphere and their contributions to global climate warming, *J. Geophys. Res.*, *116*, G02011, doi:10.1029/2010JG001393.

1. Introduction

[2] The atmospheric concentrations of major greenhouse gases (GHGs: CO₂, CH₄ and N₂O) have increased significantly since the Industrial Revolution, and have been considered a major force of climate variability and changes [Forster et al., 2007]. These three greenhouse gases in sum contribute to more than 90% of the anthropogenic climate

warming [Forster et al., 2007; Rodhe, 1990]. A host of studies have highlighted land ecosystem for its capability in absorbing CO₂ released from fossil fuel combustion, and therefore mitigating potential atmospheric and climate changes [Baldocchi, 2008; Piao et al., 2009; Schimel et al., 2001; Wofsy et al., 1993]. Despite the fact that the atmospheric concentrations of CH₄ and N₂O are much lower than that of ambient CO₂, CH₄ and N₂O highly affect the climate system with radiative efficiencies that are 25 and 298 times greater than CO₂ at a 100 year time horizon [Forster et al., 2007; Rodhe, 1990]. Compared to intensive studies on terrestrial CO₂ flux [Baldocchi, 2008; Law et al., 2002; Saleska et al., 2003; Wofsy et al., 1993], however, much less work has been done in quantifying the magnitude, spatial and temporal patterns of CH₄ and N₂O fluxes in the terrestrial ecosystems [Blais et al., 2005; Repo et al., 2009; Song et al., 2009; Zou et al., 2009].

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[3] China has drawn much attention from both the public and the scientific community as it has surpassed the United

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Figure 1. Framework showing the major components and their connections in the Dynamic Land Ecosystem Model (DLEM). (See auxiliary material for detailed information.)

States in 2006 as the largest emitter of CO_2 [Gregg et al., 2008]. A number of studies have proved that China's terrestrial ecosystems have acted as a substantial carbon sink in recent decades [Fang et al., 2007, 2001; Huang and Sun, 2006; Pan et al., 2004; Piao et al., 2009]; however, it remains uncertain whether the CO_2 sink can be translated into a mitigator for climate warming because the emissions of CH_4 and N_2O in China are relatively high [Chen et al., 2000; Ding and Cai, 2007; Huang et al., 2005, 1998, 2009; Yan et al., 2003b; Zou et al., 2010]. Thus, a comprehensive study which simultaneously examines the exchanges of CO_2 , CH_4 , and N_2O between China's terrestrial ecosystems and the atmosphere is greatly needed [Li et al., 2005; Qiu et al., 2009].

[4] Although a great number of site-specific studies on CO₂, CH₄ and N₂O fluxes have been conducted in China for several decades, few simultaneously measured all three greenhouse gases [Song et al., 2009; Tang et al., 2006; Zou et al., 2004]. At the regional scale, the spatial and temporal patterns of three biogenic GHG budgets in China remain uncertain [Qiu et al., 2009]. At present, individual technique, with either top-down or bottom-up estimate, rarely spans spatial and temporal scales to produce regional, long-term GHG budgets; furthermore, among these methods, gaps in the coverage of greenhouse gas species, emission

sources and land ecosystem types, etc. might introduce large uncertainties to the integration of multiple approaches [Schulze et al., 2009]. Process-based ecosystem models concurrently simulating CO₂, CH₄ and N₂O can be adopted as an alternative tool for investigating GHG fluxes on a regional level, as well as GHG contributions to climate change in terms of global warming potential (GWP). In this study, we used a highly integrated model, Dynamic Land Ecosystem Model (DLEM), to examine how China's terrestrial ecosystems contributed to the budgets of three GHGs during 1961–2005, and how net exchange of GHGs in terms of warming potential varied in both spatial and temporal contexts.

2. Materials and Methods

2.1. Model Description

[5] The DLEM model is a highly integrated process-based ecosystem model that aims at simulating the fluxes and storage of carbon, water and nitrogen among/within terrestrial ecosystem components while taking into consideration of multiple natural and anthropogenic perturbations. DLEM is composed of five major submodules focusing on biophysics, plant physiology, soil biogeochemistry, vegetation dynamics, land use and management as well as disturbances such as hurricane, fire, insect, etc. (Figure 1). The

fully coupled plant-soil-atmosphere continuum in model framework covers the major processes controlling uptake and release of CO₂, CH₄ and N₂O by terrestrial ecosystems. Meanwhile, the concurrent impacts of changes in climate, atmospheric composition, and land use and management practices are integrated into DLEM to retrieve reliable historical evolution and spatial patterns of the biogenic GHG budgets. DLEM has been widely applied to a variety of terrestrial ecosystem types across the globe and the detailed assumptions and processes are well documented in our previous work [Chen et al., 2006; Liu et al., 2008; Lu, 2009; Ren et al., 2007, 2011a, 2011b; Tian et al., 2008, 2010a, 2010b, 2011; Xu et al., 2010; Zhang et al., 2007].

- [6] In the DLEM model, the carbon balance of vegetation depends on carbon exchanges from photosynthesis, autotrophic respiration, and litterfall, as well as plant mortality [*Tian et al.*, 2005, 2008, 2010b]. Plants assimilate carbon via photosynthesis, and utilize portions of the absorbed carbon to compensate for the carbon loss through maintenance/growth respiration, tissue turnover, and reproduction. Net ecosystem exchange is the balance of net primary production (NPP), ecosystem heterotrophic respiration and methane flux. The NPP is the difference between gross primary production and autotrophic respiration, which have been described by *Tian et al.* [2010a].
- [7] The methane module in the DLEM model mainly simulates the production, consumption, and transport of CH₄ [Tian et al., 2010a]. Due to relatively small contribution from other substrates [Conrad, 1996; Mer and Roger, 2001], DLEM only considers the CH₄ production from dissolved organic carbon (DOC), which is indirectly controlled by environmental factors including soil pH, temperature and soil moisture content. The DOC was produced through three pathways, GPP allocation, and decomposition byproducts from soil organic matter and litterfall. CH₄ oxidation, including the oxidation during CH₄ transport to the atmosphere, CH₄ oxidation in the soil/water, and atmospheric CH₄ oxidation on the soil surface, is determined by CH₄ concentrations in the air or soil/water, as well as soil moisture, pH, and temperature. Most CH₄-related biogeochemical reactions in the DLEM were described as the Michaelis-Menten equation with two coefficients: maximum reaction rate and halfsaturated coefficient. Three pathways for CH₄ transport from soil to the atmosphere, ebullition, diffusion, and plant-mediated transport, are considered in the DLEM [Tian et al., 2010a].
- [8] In the DLEM, both denitrification and nitrification processes are simulated as one-step processes as we do not consider the midproducts in each process. Nitrification, a process converting ammonium into nitrate, is simulated as a function of soil temperature, moisture, and the NH $_4^+$ concentration [Lin et al., 2000]. Denitrification, through which the nitrate is converted into N gases, is simulated in the DLEM as a function of soil temperature, moisture, and the NO $_3^-$ concentration [Lin et al., 2000]. All the products of nitrification and denitrification that leave the system are N-containing gases. The empirical equation reported by Davidson et al. [2000] is used to separate N₂O from other gases (mainly NO and N₂).
- [9] The simulated exchanges of carbon, water and nitrogen between the interfaces at both site and regional levels have been well calibrated and extensively validated against field

observation and inventory results (see Text S1, available as auxiliary material). Especially for the three biogenic GHG budgets, simulation experiments conducted in North America and China found that DLEM performed well in capturing both the interannual/seasonal and spatial patterns of CO₂, CH₄ and N₂O fluxes [*Ren et al.*, 2007, 2011a, 2011b; *Tian et al.*, 2010a, 2010b, 2011; *Zhang et al.*, 2007]. More detailed information concerning CH₄ and N₂O production and emission can be referred to our other publications [*Tian et al.*, 2010a, 2010b; *Xu*, 2010; *Xu et al.*, 2010].

2.2. Input Data

- [10] A series of spatially and/or temporally explicit data sets have been developed to characterize multiple driving forces for the DLEM model such as changes in climate (temperature, precipitation, shortwave radiation and humidity) [Chen et al., 2006], atmospheric composition (CO₂, O₃ and N deposition) [Lu, 2009; Lu and Tian, 2007; Ren et al., 2007], and land use and management practices (fertilization, irrigation, harvest, etc.) [Liu and Tian, 2010; Liu et al., 2008] at a spatial resolution of 10 km × 10 km with a time step ranging from daily to yearly. The atmospheric CO₂ concentration data from 1900 to 2005 was obtained from the Carbon Dioxide Information Analysis Center (CDIAC, http://cdiac. ornl.gov/). The soil depth, pH, and texture maps were generated from the 1:1 million soil maps (Institute of Soil Science, Chinese Academy of Sciences) and the second national soil survey of China (National Soil Survey Office).
- [11] During 1961–2005, China's terrestrial ecosystems experienced substantial environmental changes, such as land conversion, climate change, shifts in atmospheric composition including atmospheric CO₂ concentration, ozone pollution, and nitrogen deposition (Figure 2). The atmospheric CO₂ concentration increased from 317 ppm in 1961 to 380 ppm in 2005, but we assumed that the atmospheric CO₂ concentration is spatially homogeneous across China. All the other environmental factors showed significant changing rates over time and space. We divided China into five regions: northwest, north, northeast, southwest, and southeast. The maximum, minimum, and average temperature showed a significantly increasing rate at regional and national scales; the national increasing rates are 0.29 ± 0.03 °C $(10a)^{-1}$, $0.23 \pm$ $0.04^{\circ}\text{C} (10a)^{-1}, 0.38 \pm 0.03^{\circ}\text{C} (10a)^{-1}$ for average, maximum, and minimum temperature, respectively. All the temperature changes are statistically significant for five regions. However, significance levels of precipitation change are varied among regions. Precipitation amount in the northwestern China significantly increased by 11.20 ± 3.23 mm $(10a)^{-1}$, and marginally significant increase of $28.04 \pm 16.31 \text{ mm} (10a)^{-1}$ was found in the southeastern China, while no significantly changing rate was observed for other regions; at national scale, precipitation showed a significantly increasing rate at $9.68 \pm 4.06 \text{ mm } (10a)^{-1}$ [Tian et al., 2011].
- [12] The mean monthly accumulated atmospheric ozone concentration above a threshold of 40 ppbv (AOT40) throughout the nation was increased from 100 ppb h in 1961 to 3600 ppb h in 2005, with a significantly increasing rate at 72.49 ± 3.06 ppb h yr⁻¹. The severity of ozone pollution largely varied over space and time. The increase accelerated

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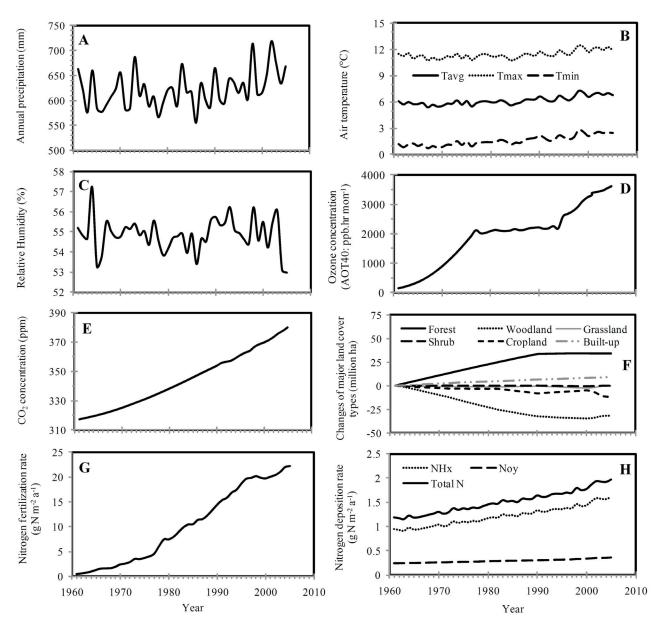


Figure 2. Temporal variation of (a) mean annual precipitation (mm), (b) maximum, minimum, and average temperature (°C), (c) relative humidity (%), (d) ozone pollution (AOT: ppb h month⁻¹), (e) atmospheric CO₂ concentration (ppm), (f) areal changes of major biome (million ha), (g) nitrogen fertilizer application rate (g N m⁻² yr⁻¹), and (h) nitrogen deposition rate (g N m⁻² yr⁻¹) along the study period.

since the early 1990s, possibly due to rapid urbanization during that period in China [*Liu et al.*, 2005a, 2005b]; over the study period, a significantly increasing rate of ozone pollution was observed for all five regions. Severe ozone pollution was observed in western and northern China, whereas southeastern and northeastern China was exposed to low-level ozone concentration (Figure 3b).

[13] The nitrogen deposition rate increased from 1.19 g N m $^{-2}$ yr $^{-1}$ in 1961 to 1.96 g N m $^{-2}$ yr $^{-1}$ in 2005, annually increasing by 16.72 \pm 0.42 mg N m $^{-2}$ yr $^{-1}$. The increasing rate of nitrogen deposition showed substantial spatial variation, with the highest annual increasing rate of 42.81 \pm 1.24 mg N m $^{-2}$ yr $^{-1}$ in southeastern China and the lowest rate of 6.21 \pm 0.34 mg N m $^{-2}$ yr $^{-1}$ in northwestern

China. The rapidest increases of both NO_y-N and NH_x-N deposition rates occurred in central and southeastern China, especially in the Yangtze River Basin, due to higher fossil fuel combustion for industrial development, N fertilizer application in cropland and winter heating [*Lu and Tian*, 2007]. In recent years, nitrogen deposition rates could have reached more than 4 g N m⁻² yr⁻¹ in recent years in southeastern China (e.g., wet deposition of 5.08 g N m⁻² yr⁻¹ in Hangzhou as measured by *Shui et al.* [1999]; wet deposition of 4.46 g N m⁻² yr⁻¹ in Dabagou and 5.63 g N m⁻² yr⁻¹ in Jinyunshan measured by EANET, 2001, http://www.eanet.cc/).

[14] The spatial and temporal patterns of nitrogen fertilizer application rate largely varied across China. The annual mean nitrogen fertilizer application increased from 0.42 g N m⁻²

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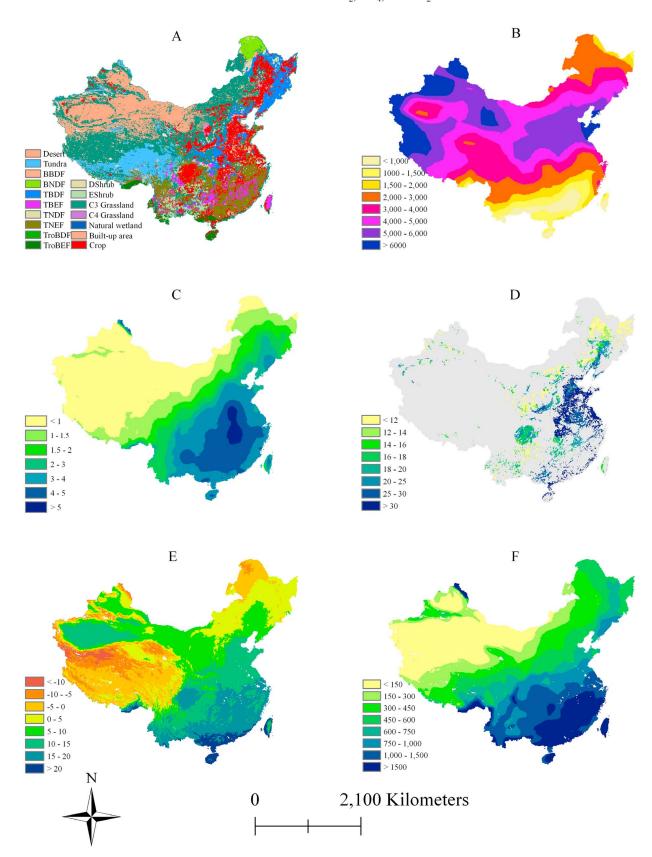


Figure 3. Spatial distribution of (a) contemporary vegetation, (b) ozone pollution (AOT: ppb h month⁻¹), (c) nitrogen deposition (g N m⁻² yr⁻¹), (d) nitrogen fertilizer application rate (g N m⁻² yr⁻¹), (e) daily air temperature (°C), and (f) mean annual precipitation (mm). (Figures 3a–3d show the data in the year of 2005, and Figures 3e and 3f show the 45 year average air temperature and precipitation.)

 ${\rm yr}^{-1}$ in 1961 to 22.23 g N m⁻² yr⁻¹ in 2005 with an overall annual increasing rate of 0.57 ± 0.01 g N m⁻² yr⁻¹ (Figure 2). The highest fertilization rates appeared in southeastern and northern China (Figure 3).

[15] According to the reconstructed historical data sets, China experienced substantial land use and land cover change during the twentieth century [Liu et al., 2005a, 2005b; Liu and Tian, 2010]. The cropland area decreased by 11.6 million ha, while built-up areas increased by 9.3 million ha and forested area showed a net increase of 34.7 million ha over the period 1961–2005. The dominant land use and land cover change in this period was land transformation from grassland and shrub to cropland and from cropland to built-up land [Ge et al., 2008; Liu et al., 2005a, 2005b]. In addition to these temporal trends, substantial spatial variations in land use and land cover change took place across China during this time period [Liu et al., 2005a, 2005b; Liu and Tian, 2010].

2.3. Model Implementation

[16] To determine the initial conditions of 1 January 1901, DLEM was fed with detrended climate data during 1901-1930. The levels of atmospheric CO₂ concentration, O₃ AOT40 index, nitrogen deposition, nitrogen fertilizer application, irrigation and land use pattern in the year of 1900 were used for the equilibrium run. A spin-up of 3000 years was applied after reaching an equilibrium state. That is, the absolute value of annual NCE (net balance of CO2 and CH4 fluxes) is less than 0.1 g C m⁻², the change in soil water pool is less than 0.1 mm, and the difference in soil mineral nitrogen content and N uptake is less than 0.1 g N m⁻² among consecutive years. Finally, the model was run in transient mode, where the simulated GHG budget was determined by the time series of multiple environmental changes from 1901 to 2005. We started the simulation in the year 1901 in order to capture the legacy effects of changes in land conversion, climate, nitrogen, O₃, and atmospheric CO₂, on greenhouse gas fluxes. This study focused on highlighting the simulation results during 1961-2005, when great changes in both natural and anthropogenic perspectives were taking place across China.

2.4. GWP Calculation

[17] The GWP index was defined to measure the time-integrated global mean radiative forcing of a pulse emission of 1 kg of a certain compound relative to that of 1 kg CO_2 [Forster et al., 2007]. According to the fourth Intergovernmental Panel on Climate Change (IPCC) report, the average GWPs for CO_2 , CH_4 and N_2O are 1, 25 and 298 at a 100 year time horizon, respectively. In this study, we applied the following equation to calculate annual GWP:

$$GWP = F_{CO_2 - C} \frac{44}{12} + F_{CH_4 - C} \frac{16}{12} \times RF_{CH_4} + F_{N_2O - N} \frac{44}{28} \times RF_{N_2O}$$

where F_{CO2-C} , F_{CH4-C} and F_{N2O-N} are annual fluxes of CO₂, CH₄ and N₂O between terrestrial ecosystems and the atmosphere based on mass of C and N, respectively. The fractions 44/12, 16/12 and 44/28 were used to convert the mass of CO₂-C, CH₄-C and N₂O-N into CO₂, CH₄ and N₂O. RF_{CH4} and RF_{N2O} are constants indicating radiative forcing of CH₄ and N₂O in terms of a CO₂ equivalent unit, and were assigned

values of 25 and 298, respectively, at 100 year time horizon [Forster et al., 2007].

2.5. Uncertainty Analysis

[18] Uncertainties in the simulated regional terrestrial fluxes of CO₂, CH₄, and N₂O were evaluated through a global sensitivity and uncertainty analysis as described by Xu [2010]. First, we assumed that the distribution of each parameter follows normal distribution based on our previous calibration experience [Tian et al., 2010a]. Second, we conducted sensitivity analysis to identify the major parameters that affect terrestrial CO₂, CH₄, and N₂O fluxes, and determine the distribution of each parameter in controlling these fluxes; the sensitivity analysis was conducted by evaluating the changes in simulated gas fluxes in response to a 20% increase and decrease of each parameter. Third, combined with the priori knowledge of parameters, we used improved Latin Hypercube Sampling (LHS) approach to randomly select an ensemble of 300 sets of 10 parameters responsible for CO₂, CH₄, and N₂O fluxes simulated by the Dynamic Land Ecosystem Model (DLEM). We set up the simulations by using the sampled 300 pairs of parameters. Finally, the outlier simulations which generated extreme fluxes for CO₂, CH₄, or N₂O will be excluded before analyzing the parameter-derived uncertainties in GHG fluxes. The 95% confidence intervals for GHG fluxes were calculated and reported.

2.6. Statistical Calculation

[19] The regression analysis was used in this study to calculate the long-term changing rates of input data and GHGs fluxes. All the statistical analyses were conducted by using the software R-program and SPSS 17.0 for Windows XP.

3. Results

3.1. National Budgets of GHGs and GWP

[20] Our model simulations indicate that China's terrestrial ecosystems acted as a net CO₂ sink with a sequestration rate of 179.3 \pm 102.2 Tg C yr⁻¹, and as a net source of CH₄ and N_2O at rates of 8.3 ± 1.5 Tg C yr⁻¹ and 0.6 ± 0.3 Tg N yr⁻¹, respectively, during 1961-2005. Over the study period, CO₂ flux in China's terrestrial ecosystems showed substantial interannual variability, ranging from a CO2 source at a rate of 48.0 Tg C yr⁻¹ in 1997 to a CO₂ sink at a rate of 415.6 Tg C yr⁻¹ in 2002; the CH₄ and N₂O fluxes also showed considerable fluctuations, the CH₄ emission could be as high as $10.4 \text{ Tg C yr}^{-1}$ in 1973 or as low as 4.1 Tg C yr^{-1} in 1978, and the N_2O emission was as high as 1.3 Tg N yr⁻¹ in 2005 and as low as 0.3 Tg N yr⁻¹ in 1966. Uncertainty analysis showed that the 95% confidence range for the fluxes of CO₂, CH₄, and N₂O are (62.0 Tg C yr⁻¹, 264.9 Tg C yr⁻¹), $(3.3 \text{ Tg C yr}^{-1}, 12.4 \text{ Tg C yr}^{-1})$, and $(0.2 \text{ Tg N yr}^{-1}, 1.1 \text{ Tg N})$ yr^{-1}), respectively (Figure 4).

[21] In terms of global warming potential, national level CO_2 uptake served to mitigate GWP at a rate of 657.5 \pm 374.9 Tg CO_2 eq yr⁻¹, while national CH₄ and N₂O emissions proved to be enhancers of GWP at rates of 275.1 \pm 48.3 Tg CO_2 eq yr⁻¹ and 285.5 \pm 131.0 Tg CO_2 eq yr⁻¹ at a 100 year time horizon. Therefore, simultaneously taking China's three biogenic GHGs into account, we found that

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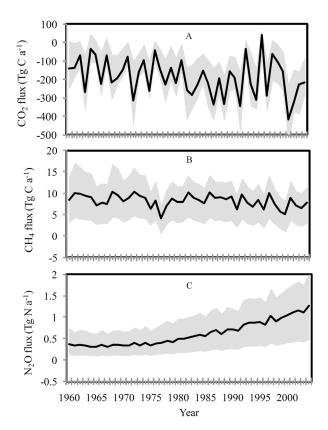


Figure 4. Temporal variations in GWP derived from fluxes of (a) CO₂, (b) CH₄, and (c) N₂O in China's terrestrial ecosystems. (Shaded area shows 95% confidence boundaries of GHGs fluxes.)

GHG-driven climate change was prone to be cooling with a GWP average of 96.9 ± 338.8 Tg CO₂eq yr⁻¹, ranging from a GWP enhancer at a rate of 766.8 Tg CO₂eq yr⁻¹ in 1997 to a GWP mitigator at a rate of 705.2 Tg CO₂eq yr⁻¹ in 2002, during the past 45 years. Our simulated results also indicate that CH₄ and N₂O emissions offset approximately 84.8% of

terrestrial CO₂ sink in China's terrestrial ecosystems for the study period.

3.2. Temporal Patterns of the GHG Fluxes and GWP

[22] Over the past 45 years, there has been a substantial interannual variation in the net exchanges of GHGs between land surface and the atmosphere across China's terrestrial ecosystems (Figure 4). Over the study period, no significant changing rate was observed for CO_2 flux, while a marginal significantly decreasing rate of 38.0 ± 15.8 Gg C yr⁻¹ ($1Gg = 10^9$ g) was observed for CH_4 emission (0.05 < P < 0.10), and a significantly increasing rate of 21.1 ± 1.1 Gg N yr⁻¹ was observed for N₂O emission (P < 0.001).

[23] We summarized the temporal patterns of the global warming potential induced by the fluxes of GHGs in Figure 5. From 1961 through 1990, a 5 year average GWP showed a slightly decreasing trend due to increased terrestrial CO₂ sequestration. However, since the 1990s, GWP resulting from biogenic GHGs began to shift from negative to positive, indicating that global radiative forcing owing to China's terrestrial GHG budgets had accumulated in this decade. The decline of CO₂ uptake in the 1990s might have been due to decreases in woodland area [*Liu and Tian*, 2010], rapid urban sprawl [*Liu et al.*, 2005a, 2005b], and climate change [*Chen et al.*, 2006]. Owing to an increased CO₂ uptake rate, GWP once again fell to negative values during 2001–2005. The lowest 5 year average net GWP occurred during 1986–1990, and the highest appeared in the period 1996–2000.

[24] Due to the broad uncertainty range of calculated GWP (Figure 5), we used 90% and 50% confidence boundaries to show the changes of GWP along the study period. The changes in GWP estimated by the DLEM is beyond the 50% confidence boundaries; for example, the upper boundary of 50% confidence boundary decreased to lower than the GWP estimated by DLEM in 1986–1990, and then increased; this indicates that it is more than 50% possibility that the reported changing pattern in GWP would occur (Figure 5).

3.3. Spatial Variations of the GHGs and GWP

[25] The biogenic fluxes of three GHGs varied substantially across China's terrestrial ecosystems. As far as CO₂ and

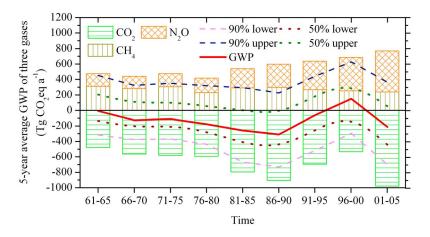
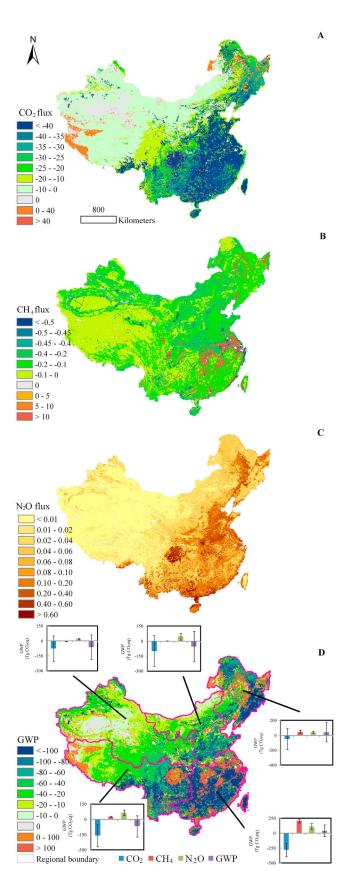


Figure 5. Temporal variations in fluxes of CO_2 , CH_4 , and N_2O and their uncertainty ranges in China's terrestrial ecosystems (90% upper and lower represent the upper and lower boundary of 90% confidence interval, respectively; 50% upper and lower represent the upper and lower boundary of 50% confidence interval, respectively; negative values indicate uptake, and positive values indicate release).



N₂O fluxes are concerned, apparent gradient was observed from southeastern China to northern and northwestern China (Figures 6a and 6c), while CH₄ emission peaked in the midsoutheast and northeast and the lowest emission or uptake was located in the central part of the mainland and northern region (Figure 6b). Over the study period, northwestern China accumulatively sequestrated 0.94 Pg CO₂-C while emitting 6.24 Tg CH₄-C and 1.83 Tg N₂O-N; northern China accumulatively sequestrated 1.21 Pg CO₂-C and oxidized 0.45 Tg CH₄-C while emitting 4.33 Tg N₂O-N; northeastern China accumulatively sequestrated 0.59 Pg CO₂-C while emitting 65.89 Tg CH₄-C and 4.12 Tg N₂O-N; southwestern China accumulatively sequestrated 1.95 Pg CO₂-C while emitting 36.22 Tg CH₄-C and 6.20 Tg N₂O-N; southeastern China accumulatively sequestrated 3.39 Pg CO₂-C, while emitting 275.94 Tg CH₄-C and 10.95 Tg N_2O-N .

[26] Among the five regions, southeastern China acted as the strongest CO_2 sink as well as the largest source of CH_4 and N_2O to the atmosphere. The overall global warming potential in this region averaged to 42.46 ± 99.21 Tg CO_2eq yr $^{-1}$ or nearly neutral over the past 45 years. It should be noted that large interannual variation existed in the contribution of biogenic GHGs from this area to the cumulative radiative forcing during 1991–2005. In terms of GWP, northeastern China, in addition to the southeastern China, is another neutral player with a rate of 43.77 ± 128.58 Tg CO_2eq yr $^{-1}$ over the past 45 years.

[27] In contrast, the other three regions exhibited a negative global warming potential with large interannual variability, which were averaged to 67.23 ± 103.97 Tg CO₂eq yr⁻¹, 61.98 ± 124.26 Tg CO₂eq yr⁻¹, and 53.96 ± 150.61 Tg CO₂eq yr⁻¹ for southwestern, northwestern, and northern China, respectively. This indicates that CO₂ uptake played a predominant role in the GHG budgets in such areas, and therefore, the terrestrial ecosystems in these three regions played an important role in alleviating global warming resulting from three major biogenic GHG fluxes. During 1961–2005, regional average GHG fluxes showed that all five regions were sinks of atmospheric CO₂ and sources of N₂O, while CH₄ uptake occurred in northwestern and northern China, and the other three regions acted as CH₄ emitters.

3.4. Biome-Level GHGs and GWP

[28] The accumulative radiative forcing due to GHG budgets in China varied among biome types. Forests acted as the strongest CO₂ sink by sequestering 3.09 Pg CO₂-C over the past 45 years, while grassland and woodland sequestered CO₂ with the orders of 1.53 and 1.01 Pg CO₂-C, respectively. Grassland was the largest CH₄ sink, accumulatively withdrawing 20.94 Tg CH₄-C during the period

Figure 6. The 45 year average of ecosystem-atmosphere exchange of (a) CO_2 (units: $g \ C \ m^{-2} \ yr^{-1}$), (b) CH_4 (units: $g \ C \ m^{-2} \ yr^{-1}$), and (c) N_2O (units: $g \ N \ m^{-2} \ yr^{-1}$), and (d) the resulted global warming potential (GWP) over China (units: $g \ CO_2$ eq $m^{-2} \ yr^{-1}$). (Note: the different scale of the y axis of insets in Figure 6d; negative values indicate uptake, and positive values indicate release; the error bars in insets represent standard deviation of GWP over the 45 years.)

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Figure 7. Contributions of various biomes to GWP over China's terrestrial ecosystem during 1961–2005. (Numbers above or below the bars are the net GWPs of three GHGs; negative values indicate cooling effect, and positive values indicate warming effect on climate system.)

1961–2005, followed by desert and others (7.75 Tg CH₄-C), forests (7.59 Tg CH₄-C), and woodland (7.58 Tg CH₄-C). It should be noted that the majority of desert in western China has a very low capacity for CH₄ oxidation due to extreme drought and lower biological activity. As for N₂O emission, the combined contribution from forest, woodland and grassland totals 10.02 Tg N₂O-N, yet is less than the N₂O released from dry cropland alone (11.94 Tg N₂O-N). Besides these three biomes, shrub land, tundra, and desert and other showed weak capacity for mitigating warming potential through sequestering CO₂ and oxidizing CH₄.

[29] When translated into GWP, forest, woodland and grassland turned out to be mitigators of climate warming (Figure 7) because the CO₂ sequestration and CH₄ oxidation-induced negative GWP in these biomes exceeded N₂O emission-induced positive GWP. China's forest played a critical role in relieving global warming, with a 45 year accumulative GWP of 10.2 Pg CO₂eq. Woodland and grassland were the second and third largest mitigators of GWP, with GWPs of 4.4 Pg CO₂eq and 2.3 Pg CO₂eq, respectively, accumulated over the study period.

[30] By contrast, accumulated GWP in both natural wetland and agricultural land (including rice paddy land and dry cropland) was positive over the past 45 years because the impacts of CH₄ and N₂O emissions on global warming were overwhelming in these ecosystems. Paddy land was the greatest contributor to national GWP due to the biogenic GHG budgets, with a 45 year cumulative value of 9.78 Pg CO₂eq, followed by dry cropland and natural wetland, in the order of 3.08 Pg CO₂eq and 2.4 Pg CO₂eq, respectively. Although both paddy land and natural wetland acted as net sinks of CO₂, sequestering 0.73 and 0.13 Pg C, respectively, during the past decades, they turned out to be contributors to GWP owing to their large emission of CH₄. From 1961 to 2005, paddy land accumulatively released 344.71 Tg CH₄-C, followed by natural wetland with a CH₄ emission

of 82.29 Tg C. In these two biomes, as *Friborg et al.* [2003] argued, CH_4 emission had a stronger effect than CO_2 sequestration on climate warming. Dry cropland ranked first in emitting N_2O to the atmosphere with total emissions of 11.94 Tg N_2O -N; its effect overweighed the cooling effects of CO_2 sequestration and CH_4 oxidation.

[31] The different contributions of various biomes to the biogenic GHG budget may explain the spatial variations of net GWP throughout China. Southeast China's role as a distinguished accelerator of global warming potential in certain years can be mainly attributed to its large proportion of rice paddies which released a great amount of CH₄. Likewise, a large area of natural wetland, which characterizes northeast China, substantially contributed to CH₄ emission. In addition, the spatial heterogeneity of net GWP may also be caused by the impacts of spatial variation in environmental factors, including climatic condition, ozone pollution, nitrogen input, and land use/cover change throughout the historical period, on biogenic GHG budgets.

4. Discussion

4.1. Comparison With Previous Studies

[32] To evaluate the reliability and robustness of our simulated results, we compared our modeled results to estimates from other studies at both site and regional levels. At the national level, China's terrestrial CO_2 uptake simulated by DLEM (198.2 \pm 113.6 Tg C yr⁻¹) is close to estimates of 186~261 Tg C yr⁻¹ for 1980–2002 [*Piao et al.*, 2009] and inversion results based on CarbonTracker (261 \pm 88 Tg C yr⁻¹) for the time period 2000–2005 (Table 1); the simulated N₂O release from China's terrestrial ecosystems is 0.71 \pm 0.17 Tg N yr⁻¹ during 1980–2000, which is slightly lower than an empirical estimate of 0.98 \pm 0.03 Tg N yr⁻¹ by *Xu et al.* [2008].

[33] Biome-level comparisons suggest that the DLEMderived estimates of net exchanges of CO₂, CH₄, and N₂O between terrestrial ecosystems and the atmosphere are consistent with other estimates based on different methods (Table 1). For example, we estimated the sink strength of atmospheric CO₂ to be 39.9 ± 32.3 Tg C yr⁻¹ for cropland, which is slightly higher yet comparable to $39 \pm 11 \text{ Tg C yr}^$ as reported by Huang and Sun [2006]; DLEM-simulated CO_2 sinks are 18.8 ± 53.1 Tg C yr⁻¹, and 3.0 ± 1.7 Tg C yr⁻¹ for grassland and natural wetlands, respectively; both are consistent with other studies (Table 1) [Duan et al., 2008; Piao et al., 2009]. DLEM-simulated CH₄ emission from natural wetland is 1.42 ± 0.50 Tg C yr⁻¹ during 1995– 2004, which is comparable to a measurement-based estimate of 1.32 Tg C yr⁻¹ [Ding and Cai, 2007]. Model simulations found that rice paddy was a net CH₄ source at a rate of $7.23 \pm$ 1.04 Tg C yr⁻¹ during 1990–2005; this value falls at the high end of the range 4.52~7.25 T g C yr⁻¹ (Table 1) [*Huang et al.*, 1998; *Wang et al.*, 2009; *Yan et al.*, 2009], but falls far lower than the estimate of 11.48 Tg C yr⁻¹ in 1991 in the work of Yao et al. [1996]. The differences between our results and previous studies could be partially explained by the different rice paddy areas accounted for; for example, Huang et al. [2006] used an area of 0.30 million km² in their estimations of CH₄ emission from China's rice paddy land in the year 2000; in contrast, we used 0.34 million km² in our study which was estimated from satellite data [Liu

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Table 1. GHGs Fluxes From Chinese Terrestrial Ecosystems Using Different Approaches^a

Reference	Piao et al. [2009]	this study	carbon tracker	this study	Piao et al. [2009]	this study	Piao et al. [2009]	this study	Duan et al. [2008]	this study	this study	this study	Ding and Cai [2007]	<i>Yan et al.</i> [2009]	Huang et al. [1998, 2006]	and Wang et al. [2009]	<i>Yao et al.</i> [1996]	this study	Xu et al. [2008]	this study	<i>Yan et al.</i> [2003a]	Xing [1998]	this study	Zheng et al. [2004]	this study	Chen et al. [2000] and	Huang et al. [2005] this study
Fluxes (T g C yr $^{-1}$ for CO $_2$ and CH4; T g N yr $^{-1}$ for N2O)	-186~-261 P	3.6	$-261 \pm 88^{\rm b}$ ca	5.4	-39 ± 11 P	-39.9 ± 32.3 th	-7 ± 2	± 53.1	-5.2^{c}	-3.0 ± 1.7 tf	8.3 ± 1.5 tt	09		5.56 Y	4.52–7.25		11.48 Y	7.23 ± 1.04 tf	0.98 ± 0.03 X	0.71 ± 0.17 th	0.476 Y	,		7	0.53 ± 0.08 th	0.0404–0.112 C	0.10 ± 0.01 df
Area (million km²) (1.2~1.6	$1.33 \sim 1.44$	3.31	3.51~3.53	0.094	$0.091 \sim 0.097$		$0.091 \sim 0.095$	0.094		0.302		0.32	0.32~0.37				0.9497	1.38		$1.37 \sim 1.40$	2.87–3.53	1.73~1.74
Period	1980–2002	1980–2002	2000–2005	2000–2005	1980–2002	1980–2002	1980–2002	1980–2002	1990s	1990s	1961–2005	1995–2004	1995–2004	2000	1995 for 1998 study and 2000	for 2006 and 2009 studies	1991	1990–2005	1980–2000	1980–2000	1995	1995	1995	1990s	1990s	$1990s^{\rm c}$	1990s
Method	inventory-based, process-based model, and inversion results	process-based model	inversion results	process-based model	inventory-based method	process-based model	inventory-based method	process-based model	field observations extrapolation	process-based model	process-based model	process-based model	site extrapolation	empirical approach	semiempirical model		regional classification	process-based model	empirical method	process-based model	empirical model	site observations extrapolation	process-based model	empirical approach	process-based model	site observations extrapolation	temperate grassland process-based model
Biome	all biomes	all biomes	all biomes	all biomes	cropland	cropland	grassland	grassland	natural wetland	natural wetland	all biomes	natural wetland	natural wetland	rice paddy	rice paddy		rice paddy	rice paddy	all biomes	all biomes	cropland	cropland	cropland	cropland	cropland	temperate grassland	temperate grassland
Gases	CO_2										CH_4								N_2O								

^aNegative values indicate uptake, while positive values indicate emission. ^bDerived from carbon tracker website (http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/). ^cBased on the field observations data used in the estimations.

Table 2. Comparison With Global Fluxes of CO₂, CH₄, N₂O, and GWP for the 1980s and the 1990s^a

	CO_2 (Pg C yr ⁻¹)	CH_4 (Tg C yr^{-1})	N_2O (Tg N yr ⁻¹)	GWP (Tg CO ₂ eq yr ⁻¹)
China	-0.19	8.44	0.70	-105.8
Global estimates	-1.95 (-2.6~-1.7) ^b	106.5 (52.5~153.75)°	9.4 (5~13.8) ^d	801.89 (-5808.57~5354.01)

^aFor gas fluxes, negative values indicate uptake, and positive values indicate release; global estimates are from Denman et al. [2007].

et al., 2005a, 2005b]. At the same time, management practices might be another reason for the estimation gaps [Li et al., 2005]. For example, Huang et al. [1998] estimated the CH₄ source in China's rice paddy land to be 7.25 Tg C yr⁻¹ in their early study, assuming a continuous flooding water regime, but later changed their estimate to 4.52 Tg C yr⁻¹ when field drainage was considered [Huang et al., 2006]. Our results show that cropland acted as a source of N₂O at a rate of 0.56 Tg N yr⁻¹ in the year 1995, which is consistent with reports by Yan et al. [2003a], Xing [1998], and Zheng et al. [2004]; DLEM-estimated N₂O source for temperate grassland is 0.10±0.01 Tg N yr⁻¹, which falls in the reported range of 0.04~0.112 Tg N yr⁻¹ during the 1990s. Therefore, it is confirmed that DLEM is capable of reasonably estimating the magnitude of terrestrial uptake and release of CO₂, CH₄ and N₂O throughout China.

4.2. Control of Interannual Variations in National GWP

[34] To further evaluate the influence of GHGs on interannual variation in national GWP, we conducted a regression analysis. A significantly positive correlation was found between annual GWP and net exchange of CO2 between terrestrial ecosystems and the atmosphere ($R^2 = 0.88$; N = 45; P < 0.001); even so, no significant correlation emerged between interannual GWP and terrestrial flux of either CH₄ or N₂O. This suggests that the interannual variability in net GWP derived from China's biogenic GHGs balance was dominated by net exchange of CO₂ between terrestrial ecosystems and the atmosphere, since CH₄ fluxes remained relatively stable and N₂O emission kept increasing, but was not potent enough to shape the net GWP over the study period. However, the contributions of CH₄ and N₂O, which determined the magnitude of GWP, were overwhelming during the warming decade of 1990-2000, during which the terrestrial CO₂ sequestration rate shrank.

4.3. Global Implications

[35] This study provides a comprehensive evaluation of the fluxes of three GHGs throughout China's terrestrial ecosystems. Putting our estimates in the context of the global fluxes of these GHGs, China's terrestrial ecosystems acted as a small mitigator for climate warming (Table 2). The summarized estimates on the fluxes of CO₂, CH₄, and N₂O in the IPCC report indicated that the global land ecosystems sequestrated CO₂ at a rate of 1.95 Pg C yr⁻¹, while emitted CH₄ and N₂O at rates of 106.5 Tg C yr⁻¹ and 9.4 Tg N yr⁻¹, respectively, over the 1980s and 1990s (Table 2). Thus, as a whole, the global land ecosystems functioned as an enhancer for climate warming at a rate of 801.9 Tg CO₂eq yr⁻¹, substantially varying from a sink at a rate of 5808.6 Tg

CO₂eq yr⁻¹ to a source at a rate of 5354.0 Tg CO₂eq yr⁻¹. In comparison, China's terrestrial ecosystems acted as a mitigator of climate warming at a rate of 105.8 Tg CO₂eq yr⁻¹ during the same time period.

4.4. Uncertainties and Improvements Needed

[36] This study, to the best of our knowledge, provides the first regional and national estimates of long-term net GWP of CO₂, CH₄, and N₂O over China's terrestrial ecosystems. Several uncertainties exist and further research is needed to improve our estimates. First, the driving forces of the model might introduce some uncertainties; for example, the area of natural wetlands (or in this case, mire) used in this study is 0.091-0.107 million km², which is close to inventoried results [Zhao, 1999], but is slightly lower than other estimates [An et al., 2007; Niu et al., 2009]. Given the considerably large difference in CH₄ flux from different wetlands [Song et al., 2009], and high uncertainty in wetland area estimates [An et al., 2007; Niu et al., 2009], there is a great need for improving data accuracy in the future. Second, intensive management on plantation forest, such as application of fertilizer, thinning, etc., was not included in the current study, and needs to be emphasized. Third, the model may oversimplify the biogeochemical and microbial processes responsible for GHGs production and consumption; for example, an atmospheric N₂O sink in soil has been reported in several field studies, but the mechanisms for N₂O sequestration in soil are still unclear [Chapuis-Lardy et al., 2007]; the inclusion of this mechanism might improve model accuracy in the estimation of national N2O flux. Fourth, the agricultural system is a highly human-dominated ecosystem which needs time series of spatially explicit data on agricultural practices across the nation for accurately estimating GHG budgets. Although our study includes irrigation, water management, harvest, fertilization and, rotation, we need to address some uncertainties associated with areas of dry cropland and rice paddies, water regime during water management, manure application, etc. [Huang et al., 2006; Zou et al., 2005].

5. Conclusion

[37] Our simulation results indicated that China's terrestrial ecosystems were a small mitigator of global warming potential in terms of three GHG budgets during 1961–2005, even though large amounts of CH₄ and N₂O were released during this period. Approximately 84.8% of the cooling effects induced by terrestrial CO₂ sink were offset by terrestrial CH₄ and N₂O emissions in China. We also found that large interannual variations in biogenic GHG contributions to climate change existed on national scale. Southeast and northeast China were the neutral areas contributing little to

^bLand ecosystems only [Denman et al., 2007].

^cEmission from wetland and soil consumption of atmospheric CH₄.

^dSource from both agricultural and natural soils across the globe.

warming potential over the past 45 years, even though GWP values in these two regions shifted significantly from negative to positive since 1990. By contrast, the accumulated negative radiative forcing due to prevailing terrestrial CO₂ uptake in northwest, north, and southwest China considerably reduced the impact of GHGs on climate warming. Paddy land was the greatest contributor to national GWP, followed by natural wetland and dry cropland. China's forest area played a critical role in alleviating warming potential, with grassland and woodland as the second and third mitigators. Comparing to global terrestrial ecosystems acting as an enhancer of climate warming, China's land ecosystems functioned as a mitigator of climate warming for the past 45 years.

[38] Studies on concurrent net exchanges of greenhouse gases between land surface and the atmosphere, rather than a certain GHG species, would provide more profound insights for scientific communities and policy makers striving to curb climatic warming. We are calling for more site-level experiments highlighting fluxes of multiple GHGs and their interactions to validate process-based model results, and extrapolate their underlying mechanisms to wider ranges. In addition to accurately quantifying GHG budgets, future work is greatly needed to investigate the contributions of multiple environmental changes which control global warming potential by affecting GHG budgets.

[39] Acknowledgments. This study has been supported by NASA LCLUC Program (NNX08AL73G_S01), NASA IDS Program (NNG04GM39C), and China's Ministry of Science and Technology (MOST) 973 Program (2002CB412500). We thank Yao Huang in Institute of Atmospheric Physics, Chinese Academy of Sciences (CAS), Xiaoyuan Yan in Institute of Soil Science, CAS, and all members of the Ecosystem Dynamics and Global Ecology (EDGE) Lab for their helpful comments and discussions. We also appreciate Dennis Baldocchi and two anonymous reviewers for their constructive comments which greatly improved this manuscript.

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