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Nitrogen deposition: how important is it for global terrestrial carbon uptake?

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Abstract

Global carbon budget studies indicate that the terrestrial ecosystems have remained a large sink for carbon despite widespread deforestation activities. CO_2-fertilization, N deposition and re-growth of mid-latitude forests are believed to be key drivers for land carbon uptake. In this study, we assess the importance of N deposition by performing idealized near-equilibrium simulations using the Community Land Model 4.0 (CLM4). In our equilibrium simulations, only 12–17 % of the deposited Nitrogen is assimilated into the ecosystem and the corresponding carbon uptake can be inferred from a C : N ratio of 20 : 1. We calculate the sensitivity of the terrestrial biosphere for CO_2-fertilization, climate warming and N deposition as changes in total ecosystem carbon for unit changes in global mean atmospheric CO_2 concentration, global mean temperature and Tera grams of Nitrogen deposition per year, respectively. Based on these sensitivities, it is estimated that about 242 PgC could have been taken up by land due to the CO_2 fertilization effect and an additional 175 PgC taken up as a result of the increased N deposition since the pre-industrial period. Because of climate warming, terrestrial ecosystem could have lost about 152 PgC during the same period. Therefore, since preindustrial times terrestrial carbon losses due to warming may have been approximately compensated by effects of increased N deposition, whereas the effect of CO_2-fertilization is approximately indicative of the current increase in terrestrial carbon stock. Our simulations also suggest that the sensitivity of carbon storage to increased N deposition decreases beyond current levels, indicating climate warming effects on carbon storage may overwhelm N deposition effects in the future.

1 Introduction

Though nitrogen is the most abundant element in the atmosphere, most organisms including plants and animals cannot use it in its most common form (N_2). It can be used only in reactive forms of NO_y and NH_x, which when deposited on the surface through various processes is generally referred to as the "nitrogen deposition". Since the prehistoric time, nitrogen has been converted to bioavailable forms through "lightening", bacteria, algae, legumes and plants with associative N_2 fixers. The rate of nitrogen fixation (and thus deposition) has significantly increased due to systematic cultivation of rice and leguminous plants. The rate is estimated to be ~ 10.8 $Tg N yr^{-1}$ (Tera grams of Nitrogen per year) around 1765 (Galloway et al., 2004; Jain et al., 2009) and around 17.4 $Tg N yr^{-1}$ by 1865 (Galloway et al., 2004).

In the industrial era, nitrogen deposition has increased many times due to nitrogenous emissions from fossil fuel combustion and due to industrial production of various reactive nitrogen compounds from Haber–Bosch process. As a result, N deposition increased to about 60 $Tg N yr^{-1}$ in the 1990s (Galloway et al., 2004; Jain et al., 2009) and is projected to increase to about 125 $Tg N yr^{-1}$ by 2050 (Galloway et al., 2004). Estimates of N deposition based on NO_y emissions alone are between 25 and 40 $Tg N yr^{-1}$ for the year 2000 and 60–100 $Tg N yr^{-1}$ by 2100 (Lamarque et al., 2005).

Global carbon budget studies suggest that the global terrestrial ecosystem remains a large sink of carbon in recent decades despite a widespread deforestation related flux of carbon to the atmosphere (Friedlingstein et al., 2010; Le Quere et al., 2009; Pan et al., 2011; Schimel, 1995). Regrowth of mid-latitude forests, CO_2-fertilization and nitrogen deposition are believed to be key drivers for land carbon uptake (Canadell et al., 2007; Friedlingstein et al., 2010; IPCC, 2007). It is well documented that CO_2 fertilization increases carbon fluxes into land and ecosystem carbon stocks. For example, free-air CO_2 enrichment experiments in forest (Norby et al., 2005) stands indicates a 23 % median increase in net primary production (NPP) in response to a CO_2 concentration increase from 376 to 550 ppm (parts per million). Warming in contrast is known to diminish the land carbon sink by enhancing respiration (Cox et al., 2000; Cramer et al., 2001; Friedlingstein et al., 2001; Govindasamy et al., 2005; Joos et al., 1991; Lloyd and Taylor, 1994; Matthews et al., 2005; Thompson et al., 2004; Zeng et al., 2004). Global mean temperature is estimated to have increased by about 0.74 °C in the period 1906 to 2005 (IPCC, 2007).

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A number of observational studies from different geographical areas have shown that N deposition increases carbon stocks in different plant species (see Table S1 for a brief list of studies and their results). The amount of additional carbon stock increase depends primarily on the C : N ratio of the ecosystems: an estimate of upper bound for present day would be 1–2 $Pg\,C\,yr^{-1}$ for a C : N ratio of 20–40 : 1 and 50 $Tg\,N\,yr^{-1}$ increase in N deposition relative to pre-industrial period. Beyond certain N deposition levels additional N deposition has reduced impact on biomass yield and productivity (Lemus et al., 2008; Rasmussen, 1998). While modeling studies show that N deposition increases NPP and carbon stocks (Jain et al., 2009; Magnani et al., 2007; Thornton et al., 2009; Yang et al., 2009), the importance of N deposition relative to CO_2 fertilization effect and temperature increases has not been adequately explored on a global scale. A recent coupled modeling study shows N deposition and elevated CO_2 could have synergistic effect which could explain 47 % of terrestrial carbon uptake in the 1990s (Churkina et al., 2009). Estimates of global terrestrial carbon uptake due to current N deposition range from 0.15–0.35 $Pg\,C\,yr^{-1}$ (10–20 % of terrestrial uptake) (de Vries, 2009; de Vries et al., 2008; Zaehle and Dalmonech, 2011) to 1.0–2.0 $Pg\,C\,yr^{-1}$ (100 % of terrestrial uptake) (de Vries, 2009; de Vries et al., 2008; Holland et al., 1997; Magnani et al., 2007; Zaehle and Dalmonech, 2011) or 0.31 $Pg\,C\,yr^{-1}$ in tree carbon storage (Thomas et al., 2010). However, there are indications that N-induced increase in land carbon uptake is unlikely to keep pace with future CO_2 increases (Reay et al., 2008).

Terrestrial carbon accumulation could be constrained by the availability of nitrogen (Hungate et al., 2003; Nadelhoffer et al., 1999) and because of this constraint it has been found (Bonan and Levis, 2010; Jain et al., 2009; Thornton et al., 2009; Yang et al., 2009) that nitrogen cycle dynamics attenuates the magnitude of global terrestrial carbon sinks and sources driven by CO_2 fertilization and changes in climate. However, these studies do not provide a quantitative estimate of how much terrestrial carbon stock is increased by unit nitrogen deposition. Further, how this sensitivity to N deposi-

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tion will change under climate warming and changing atmospheric CO_2 concentration is not yet investigated. In this study, we address the following three questions:

1. How much carbon could be sequestered into terrestrial ecosystem per $Tg\,N\,yr^{-1}$ increase in N deposition?

2. How does the sensitivity to N deposition respond to the changing temperature and CO_2 concentration?

3. What is the importance of N deposition relative to CO_2-fertilization and global warming in determining total ecosystem carbon storage? (Total ecosystem carbon (TEC) is the sum of all terrestrial carbon pools in vegetation, soil and litter.)

To address these issues, we use a global land model coupled to carbon and nitrogen cycles. Our simulations are highly idealized since our main goal here is to get an order of magnitude estimate for the sensitivity of terrestrial ecosystem to N deposition, climate warming and CO_2-fertilization. Further, we design near-equilibrium simulations as opposed to transient simulations since there could be substantial lags in terrestrial ecosystem response (Jones et al., 2009) and equilibrium simulations allow us to capture long term consequences. However, it should be cautioned that the sensitivity parameters estimated from equilibrium simulations have much larger magnitudes when compared in transient simulations as shown in one of our recent studies (Bala et al., 2012). Though our simulations are highly idealized the results may have important implications for the terrestrial carbon dynamics for the historical and future periods.

2 Model description

To investigate the relative influence of N deposition, CO_2 fertilization and climate warming on ecosystem carbon productivity, we use the Community Land Model CLM4 (Lawrence et al., 2011). CLM4 merges the biophysical framework of the CLM 3.5 (Oleson et al., 2008, 2010; Stockli et al., 2008) with the terrestrial biogeochemistry model Biome BGC (version 4.1.2) (Thornton and Rosenbloom, 2005; Thornton et al., 2002). CLM4 includes revised hydrology and snow models, organic soils, and a 50 m deep ground column when compared to CLM3.5. Additionally in CLM4 the distribution of plant functional types (PFTs) is modified to reduce a high grass bias in forested regions. It includes carbon-nitrogen biogeochemistry with prognostic carbon and nitrogen in vegetation, litter, and soil organic matter (Thornton and Zimmermann, 2007; Thornton et al., 2009). A prognostic fire model simulates wild fires (Kloster et al., 2010). In CLM4, Nitrogen input to the ecosystem is biological fixation and N deposition. Within the ecosystem, Nitrogen is released from organic matter (gross mineralization) in forms that can then be taken up by plants (plant uptake or assimilation) and remaining is immobilized (Immobilization). N losses from ecosystem are through fire loss, denitrification and leaching.

CLM4 is forced by a 57 yr (1948–2004) observationally constrained atmospheric forcing dataset at a three-hourly intervals for surface air temperature, precipitation, surface pressure, boundary layer wind and surface solar radiation at a horizontal resolution of 1.9° latitude and 2.5° longitude (Qian et al., 2006). Inputs to the model such as, the initial conditions, the surface parameters and the plant functional type physiological constants were all set from the input dataset associated with distribution of CLM4 source code. The 15 PFTs that are prescribed in the model corresponds to present-day vegetation cover and land cover and land use change is not considered. Prescribed constant level of atmospheric CO_2 concentration forcing is used for each simulation.

3 Experiments

We performed twelve 1000 yr simulations with the same climate forcing but varying N-deposition, CO_2 concentrations and climate warming over the globe to isolate the effects of these factors on global ecosystem productivity and carbon storage. The twelve experiments are grouped into 3 sets as follows: (1–4) 1N (Control), 2N, 4N and 8N where atmospheric CO_2 concentration is fixed at the pre-industrial (285 ppm;

year 1850) levels and N deposition is 1x, 2x, 4x and 8x the pre-industrial levels, respectively, (5–8) 1N2xCO2, 2N2xCO2, 4N2xCO2 and 8N2xCO2 are same as (1–4) but the CO_2 level is doubled, (9–12) 1N2K, 2N2K, 4N2K and 8N2K are same as (1–4) but a uniform increase of 2K in atmospheric temperature forcing is imposed. All twelve simulations start from a spun up pre-industrial state and changes in N deposition, CO_2 and climate warming are imposed as step-function changes at the start of the simulations. The 57 yr atmospheric forcing dataset is repeatedly used in all twelve 1000 yr experiments.

The prescription of nitrogen deposition in our simulations is designed so as to capture the pre-industrial, current and projected future nitrogen deposition levels on the global land system: N deposition in 1N (preindustrial period) and 2N are prescribed at 20.3 $TgNyr^{-1}$ (Bonan and Levis, 2010) and 40.6 $TgNyr^{-1}$, respectively, over land whereas the present day N deposition is 65.2 $TgNyr^{-1}$ in CLM4 dataset. The N deposition used in CLM4 (Fig. 1) were generated by the three-dimensional chemistry transport MOZART-2 (Model for Ozone and Related Tracers, version 2, Horowitz et al., 2003). These N deposition levels are close to the values reported in literature i.e 17.4 TgN yr^{-1} in 1860 and 62 $TgNyr^{-1}$ in the year 2000 (Galloway et al., 2004; Jain et al., 2009). The prescribed N deposition over land in the experiment 8N is 162.4 $TgNyr^{-1}$ while the projected N deposition in 2050 is 135 $TgNyr^{-1}$ (Galloway et al., 2004).

While the first set of experiments (1–4) is designed to estimate the response of the model to N deposition, the second set of experiments (5 to 8) is designed to estimate the sensitivity of the model to CO_2-fertilization and the interaction between CO_2-fertilization with N deposition. The third set of experiments (9 to 12) is designed to calculate the sensitivity to climate warning and its interaction with N deposition.

4 Results

The spatial pattern of N deposition used in our experiments based on pre-industrial N deposition is similar to present day deposition (Fig. 1). South and Southeast Asia,

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Europe, Eastern North America and Central Africa have larger N deposition. The pattern of deposition is primarily determined by sources of reactive nitrogen inputs to the atmosphere, atmospheric transport and wet and dry deposition processes in the atmosphere (Horowitz et al., 2003).

The changes in key terrestrial carbon cycle variables (Net Primary Productivity (NPP), vegetation carbon, soil carbon and total ecosystem carbon) for elevated N deposition are shown in Fig. 2 which shows that the simulations have reached near-equilibrium conditions after 900 yr and hence we use the last 100 yr in our analysis. During the last 100 yr period, net ecosystem exchange (NEE) has a magnitude between 0.01 and 0.1 $\mathrm{Pg\,C\,yr^{-1}}$ in all the simulations. Figure 2 suggests that as the N deposition increases, NPP, vegetation carbon, soil carbon and TEC also increase. It demonstrates the presence of N limitation in the terrestrial ecosystems (Vitousek and Howarth, 1991) as addition of N deposition results in increased NPP. It also suggests that at lower N deposition levels the terrestrial ecosystem is more sensitive to addition of nitrogen and is less sensitive at higher N deposition levels. Further, we find that climate warming leads to a decrease in TEC and the decrease is larger when N deposition levels are larger. The causes for the dependence of sensitivity on N deposition levels are discussed in the following paragraphs.

Figure 3 and Table 1 show that the simulated TEC averaged over the near-equilibrium period (900–1000 yr) increases substantially as N deposition rate is increased. The increases are 69 PgC (3.6 %), 183.5 PgC (9.4 %), and 352 PgC (18.1 %) for doubling, quadrupling and eight times N deposition, respectively. TEC increases per $\mathrm{Tg\,N\,yr^{-1}}$ are 3.41, 3.01, and 2.48 $\mathrm{Pg\,C\,(Tg\,N\,yr^{-1})^{-1}}$ for these three cases, respectively. That is, TEC increase per unit N deposition becomes smaller for large N deposition, and eventually the system would reach steady state and thus the land biosphere will eventually stop being a carbon sink (Rasmussen, 1998). The sensitivity for N deposition decreases at higher N deposition levels because biological nitrogen fixation (BNF; an input of N to terrestrial ecosystem) saturates at higher NPP as formulated in

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CLM4 (Oleson et al., 2010):

$$BNF = 1.8(1 - \exp[-0.003\ NPP])$$

The saturation of BNF at higher NPP (associated with higher N deposition) is intended to represent the hypothesis that N fixation is eventually limited by other nutrients, especially phosphorus.

An exponential fit with 2 time constants shows that major changes in TEC occur on decadal and centennial time scales for step function changes in N deposition, temperature and CO_2 (Table S2). Therefore, on centennial time scales, the order of magnitude TEC increase for N deposition can be inferred from accumulation of total ecosystem nitrogen (TEN; Table 1) due to N deposition. For an average C : N ratio of 20 : 1 for the total terrestrial ecosystem (approximate ratio of TEC to TEN in Table 1), when N deposition is increased by 20.3 Tg N yr^{-1} (2N-1N), we find an increase in TEN of 3.4 Pg N and an associated TEC increase of 69 PgC. Our model-based estimate is conservative when compared to observations in European sites which find a carbon sequestration range of 5–75 kg C per kg N for forests and heartlands and a most common range of 20–40 kg C kg^{-1} N^{-1} (de Vries et al., 2009) or US sites which find above-ground biomass increment of 61 kg of carbon per kg of nitrogen deposited (Thomas et al., 2010). Defining an overall N-accumulation fraction as the ratio of ecosystem N-accumulation to N deposition for the entire period (1000 yr), we get an N-accumulation fraction in the range of 12–17 % (12 % for 8N-1N and 17 % for 2N-1N). Therefore, for our equilibrium simulations only 12–17 % of the deposited Nitrogen is assimilated into the ecosystem and the corresponding carbon uptake can be inferred from a C : N ratio of 20 : 1.

Figure 3 also shows that under the warming scenario, the simulated TEC averaged over the years 900–1000 declines for all N deposition levels. The TEC decreases for a 2K warming by 303.4 (15.6 %), 315.8 (15.7 %), 335.5 (15.8 %) and 365.1 (15.9 %) Pg C, respectively, at preindustrial level N deposition (1N2K-1N), 2 times (2N2K-2N), 4 times (4N2K-4N) and 8 times (8N2K-8N) the preindustrial N deposition levels. While

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the absolute magnitudes of these changes show an increase with the background N deposition levels, the similar percentage changes suggest that the sensitivity of TEC to warming remains almost a constant for the levels of N deposition considered in this study. This indicates a "pool size" effect: at higher N deposition levels, the carbon stocks are higher and hence the change per unit warming is larger though the percentage change is constant.

The CO_2-fertlization leads to an increase in TEC at all levels of N deposition (Fig. 3). The simulated TEC averaged over the years 900–1000 increases for a doubling of CO_2 by 627.7 (32.3 %), 649.5 (32.2 %), 689.9 (32.4 %) and 758.4 (33 %) Pg C respectively, at preindustrial level N deposition (1N2xCO2-1N), 2 times (2N2xCO2-2N), 4 times (4N2xCO2-4N) and 8 times (8N2xCO2-8N) the preindustrial N deposition levels. The percentage changes suggest that the sensitivity of TEC to CO_2 fertilization also remains almost a constant for the levels of N deposition considered in this study. However, the absolute magnitudes show an increase with the background N deposition levels as was the case with warming, indicating the "pool size" effect identified above.

Spatial pattern of changes in TEC under different N deposition levels and under warming and CO_2 fertilization levels for the last 100 yr of simulations are shown in Fig. 4. Overall, N deposition leads to enhanced TEC (Fig. 4) as increased N deposition leads to increase in BNF and N-fixation (Fig. 5). We also find that climate warming leads to a decline in TEC except in northern high latitudes (Fig. 4) where warming results in longer growing season and increased TEC. CO_2-fertlization causes an increase in TEC everywhere with centers of maxima seen in the Amazon, central Africa and Southeast Asia (Fig. 4). Climate warming and CO_2-fertilization lead to decrease and increase, respectively, in total ecosystem nitrogen which are primarily driven by changes in denitrification, BNF and fire loss nitrogen (Fig. 6).

The feedbacks of terrestrial biosphere to increasing CO_2 concentration and warming have been well quantified (Bala et al., 2012; Boer and Arora, 2009; Friedlingstein et al., 2003, 2006; Zickfeld et al., 2011) using two parameters that determine the land carbon uptake: the carbon storage sensitivity over land to CO_2 (β_L; beta) and to temperature change (γ_L; gamma). In this study we introduce a new sensitivity parameter i.e. carbon storage sensitivity over land to N deposition (δ_L; delta) to quantify the response for N deposition. β_L is defined (Bala et al., 2012; Friedlingstein et al., 2003; Friedlingstein et al., 2006) as the change in TEC associated with unit change in atmospheric CO_2 (C_a), γ_L as the change in TEC associated with unit change in temperature and δ_L as the change in TEC associated with unit change in atmospheric N deposition:

$$\beta_L = \frac{\Delta \text{TEC}}{\Delta C_a} \quad (1)$$

$$\gamma_L = \frac{\Delta \text{TEC}}{\Delta T} \quad (2)$$

$$\delta_L = \frac{\Delta \text{TEC}}{\Delta N} \quad (3)$$

C_a,T and N refer to atmospheric CO_2 concentrations, global-mean surface temperature and atmospheric N deposition rate to soil mineral Nitrogen.

Table 2 shows the values of β_L, γ_L and δ_L and the time evolution of these parameters are shown in Fig. 7. We find that CO_2 fertilization (β_L) and N deposition (δ_L) lead to increases in TEC. TEC increases $\sim 2.1\,\text{Pg C ppm}^{-1}$ in response to increased atmospheric CO_2 concentration, and by $\sim 3.4\,\text{Pg C}\,(\text{Tg N yr}^{-1})^{-1}$ in response to increased N deposition. However warming causes TEC to decrease by $\sim 152\,\text{Pg C K}^{-1}$. Table 2 also suggests that with the increasing terrestrial N deposition, the magnitude of TEC sensitivity to CO_2 fertilization increases as does the negative TEC sensitivity to warming due to the "pool size" effect discussed earlier. Further, we find that TEC sensitivity to N deposition decreases with increasing N deposition levels and it increases (decreases) in the presence of CO_2-fertilization (climate warming). The equilibrium values of β_L and γ_L in our simulations also are larger (Bala et al., 2012) when compared with previous transient CCSM simulations and stand-alone-land model simulations (Bonan and Levis, 2010; Thornton et al., 2009). For a doubling of CO_2, the TEC increases by

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32.3 % after 900 yr of stabilization which is close to the 28 % increase found by a recent study (Bala et al., 2012) for a doubling CO_2 in a coupled climate model.

Figure 8 shows that β_L is positive at all land points and β_L shows a slight increase as N deposition levels increase (Table 2). γ_L decreases in most regions (Fig. 8) because climate warming results in reduced NPP and the consequent declines in vegetation and soil carbon. However, in the northern high latitudes warming leads to higher ecosystem productivity and hence positive γ_L values. At higher levels of N deposition concentrations, unit increase in temperature results in larger ecosystem carbon losses (Table 2). At lower levels of N deposition concentration, an increase in N deposition results in larger ecosystem carbon increase – tropical and temperature regions show relatively large increases in TEC as N deposition increases from 1N to 2N (Fig. 8). However at higher levels of N deposition, increases are moderate. Also TEC sensitivity to N deposition decreases for present day deposition when compared to pre-industrial N deposition levels (Fig. S1). This shows that the magnitude of TEC sensitivity to N deposition is likely to decrease beyond current N deposition levels.

We perform an additional simulation (1NPREC2K) to investigate effects of hydrological cycle changes, because in our climate warming simulations we have imposed only temperature changes but not the associated changes in other important variables such as precipitation, water vapor and clouds. In 1NPREC2K, we imposed a uniform increase in precipitation of 6 % and specific humidity increase of 13 % in association with the 2 K warming as global mean precipitation and specific humidity are constrained to increase by ~3 % and 6.5 % per unit warming, respectively (Allen and Ingram, 2002; Held and Soden, 2006). A comparison of spatial pattern of changes in TEC in 1NPREC2K and 1N2K indicates that the experiment 1N2K without the climate change related precipitation and water vapor changes is able to simulate the TEC changes associated with a 2K global mean warming very well (Fig. S2) as regional differences in TEC between 1NPREC2K and 1N2K are at most only ~ 10–15 %. Further, increased N deposition could potentially affect the hydrological cycle by increasing the leaf area index and canopy transpiration. However, we find that the simulated effect of N deposition on land hydrological cycle is much smaller when compared to effects from CO_2-fertlization and climate warming (Fig. S3).

Finally, we assess if there is any nonlinearity (or two-way interaction) in our simulations (Table S3). We find that the combined effect for warming and N deposition is approximately close to the sum of individual effects at smaller N deposition levels indicating the near-absence of two-way interactions. However, larger deviations from linearity appear at larger N deposition. For instance, at eight times the pre-industrial N deposition levels the difference between combined effect and sum of effects is about 62 PgC^{-1}. The interaction between climate change and N deposition imply a loss of TEC and the sign of net TEC change is altered by the interaction (Table S3). Similar near-linearity at small N deposition and significant nonlinearity at higher N deposition can be seen for the combination of CO_2-fertilization and N deposition effects (Table S3). In this case, the interaction implies a gain of TEC for ecosystems. The negative sign of the two-way interaction for climate change and positive sign for CO_2-fertilization are merely a reflection of the fact that the TEC sensitivity to N deposition (δ_L) is larger under CO_2-fertilization and smaller under climate change (Table 2).

5 Discussion and conclusions

What are the key drivers of the terrestrial carbon uptake in the recent decades? While the role of carbon fertilization and climate warming is well studied, the role of nitrogen deposition remains under explored. N deposition has increased from 10.8 TgN in 1765 to 62.2 TgN in 2000 (Galloway et al., 2004; Jain et al., 2009). During the same period, the atmospheric CO_2 has increased by about 110 ppm and the global mean temperature has increased by about 1 K (IPCC, 2007). Our analysis of the TEC sensitivity to CO_2 fertilization (β_L) and N deposition (δ_L) suggests that about 242 PgC could have been taken up by land due to the CO_2 fertilization effect and an additional 175 PgC taken up as a result of the increased N deposition since the pre-industrial period. Because of climate warming (γ_L), terrestrial ecosystem could have lost about 152 PgC

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during the same period. The zonal mean percentage changes in TEC due to these three factors show similar orders of magnitude (Fig. 9, right panels). We caution that our estimates provide only an order of magnitude of the three effects considered in this study since our simulations are idealized near-equilibrium simulations. We are justified in using near-equilibrium sensitivity values for the transient historical period since major TEC changes occur on decadal and centennial time scales (Table S2).

These estimates indicate that TEC losses due to increased warming are likely more than compensated by the additional N deposition since the pre-industrial period (Fig. 9). The land biosphere has been a sink for carbon because N deposition and warming impacts approximately cancel each other while CO_2-fertilization effect is feeding the current increase in ecosystem productivity. While the contribution of CO_2-fertilization and warming to TEC are well known, our study suggests N deposition to be an equally important factor controlling the terrestrial carbon cycle. There have been indeed suggestions that terrestrial carbon loss due to deforestation and agriculture may have been more or less balanced by nitrogen-stimulated carbon uptake (Schindler and Bayley, 1993). The N deposition is projected to increase by about 8 times by 2050s relative to preindustrial levels (Galloway et al., 2004). Our analysis suggests that as N deposition increases the sensitivity of TEC to N deposition decreases (Fig. 7) due to two factors. First, for a constant N-deposition rate the annual increase in TEC decreases with time (see the exponential fit for TEC changes in Table S2). Second, for a specified amount of increase in N deposition the increase in TEC decreases with the amount of pre-existing N deposition. Both of these factors would lead to a decrease in the magnitude of TEC increase over time. Therefore, it is likely that increasing N deposition may not be able to compensate the loss in TEC caused by warming in the future.

Our findings should be viewed in the light of the limitations and uncertainties involved in this study. One of the key limitations is that we have used an offline version of CLM and hence the feedbacks with other components of the climate system (e.g. atmosphere and ocean) are missing in our simulations. However, our results should

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not differ substantively from those obtained with more comprehensive models, and use of a simpler model permits isolation of effects of different causal factors (i.e., CO_2 level, temperature, and amount of N deposition). For instance, our present analysis suggests that TEC could increase by 628 PgC (32.3 %) for a doubling of CO_2 which is in close agreement with a recent study (Bala et al., 2012) which found 28 % increase for a doubling of CO_2 in a coupled model that had CLM as its land model component.

In our climate change experiments, we have not considered land use and land cover change. The radiative effect of N_2O emissions associated with N deposition is also not included in this study. There are indications that the C-sink benefit offered by N deposition could be significantly offset by the warming potential of associated N_2O emissions (Dolman et al., 2010; Reay et al., 2008; Xu et al., 2012). Recent studies (Tian et al., 2012) do indicate that the warming effect associated with N_2O emissions in "over fertilized" regions has completely counteracted the carbon sink effect in some regions of the world. Our model has neither the representation for Ozone (produced by elevated NO_x) damage to plants (Krupa et al., 2001) nor the NPP and ecosystem carbon decline due to soil acidification from sustained Nitrogen deposition (Rasmussen, 1998).

Our study is an idealized modeling study which investigates the near-equilibrium changes and does not quantify the changes from transient forcing. Therefore, it is likely that the magnitudes of the sensitivity parameters estimated in this study are larger than would be obtained in transient simulations (Bala et al., 2012). This study is based on a single model CLM which is one of a few models with representations for both carbon and nitrogen cycles. Our understanding of nitrogen cycle and carbon-nitrogen interaction is weak and has major uncertainties (Dolman et al., 2010; Reay et al., 2008; Zaehle and Dalmonech, 2011) and hence more observational and modeling studies especially multi-model intercomparsions will be required to provide more confidence.

Increased atmospheric CO_2 and increased N deposition both increase carbon storage in terrestrial ecosystems. In contrast, increased temperatures decrease terrestrial carbon storage. Our model results suggest that over past and future decades, human-induced changes in N deposition are of the same magnitude but opposite in sign to effects of human-induced temperature changes on terrestrial carbon storage. Thus, the increase in terrestrial carbon stock is likely to be the same magnitude as the effect of CO_2 fertilization on this stock. However, our results indicate that the effectiveness of N deposition in increasing terrestrial carbon storage is likely to decrease as time goes on, and thus temperature effects are likely to ultimately overwhelm effects of increased N deposition. Nevertheless, effects of increased atmospheric CO_2 concentrations are likely to dominate the overall response leading to increased total ecosystem carbon storage.

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/10/11077/2013/bgd-10-11077-2013-supplement.pdf.

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Table 1. Global mean changes in key ecosystem variables in the last 100 yr of 1000 yr simulations. Values in parenthesis are % changes.

Key Terrestrial Variables	1N	Set 1 Experiments: Increasing N deposition alone			Set 2 Experiments: Increasing N deposition with 2 K warming				Set 3 Experiments: Increasing N deposition with doubled CO_2 concentration			
		2N-1N	4N-1N	8N-1N	1N2K-1N	2N2K-2N	4N2K-4N	8N2K-8N	1N2xCO2-1N	2N2xCO2-2N	4N2xCO2-4N	8N2xCO2-8N
GPP (Pg C yr^{-1})	184.4	6.2 (3.4)	16.4 (8.9)	30.8 (16.7)	−17.4 (−9.4)	−18.4 (−9.7)	−20.0 (−10.0)	−22.2 (−10.3)	53.8 (29.2)	56.0 (29.4)	60.0 (29.9)	66.8 (31.0)
NPP(Pg C yr^{-1})	63.8	2.6 (4.0)	6.8 (10.6)	12.8 (20.1)	−8.0 (−12.6)	−8.4 (−12.7)	−9.1 (−12.9)	−10.1 (−13.2)	18.0 (28.2)	18.8 (28.3)	20.3 (28.7)	22.8 (29.7)
Vegetation Carbon (Pg C)	1066.8	28.0 (3.0)	45.6 (4.7)	65.4 (6.45)	−179.0 (−19.0)	−185.7 (−19.2)	−195.8 (−19.3)	−210.5 (−19.5)	432.8 (40.6)	445.3 (40.5)	468.9 (40.8)	508.3 (41.5)
Vegetation Nitrogen (Pg N)	4.9	0.2 (3.5)	0.5 (9.1)	0.9 (17.3)	−0.7 (−14.8)	−0.8 (−14.9)	−0.8 (−15.0)	−0.9 (−15.3)	1.6 (31.9)	1.6 (32.0)	1.7 (32.4)	1.9 (33.3)
Soil Carbon (Pg C)	743.8	32.2 (4.3)	86.5 (11.6)	168.1 (22.6)	−81.5 (−11.0)	−85.6 (−11.0)	−92.5 (−11.1)	−103.6 (−11.4)	151.1 (20.3)	158.8 (20.5)	172.7 (20.8)	196.9 (21.6)
Soil Nitrogen (Pg N)	74.2	3.2 (4.3)	8.6 (11.6)	16.8 (22.6)	−8.1 (−11.0)	−8.5 (−11.0)	−9.2 (−11.1)	−10.3 (−11.4)	15.1 (20.3)	15.8 (20.5)	17.2 (20.8)	19.7 (21.6)
Total Ecosystem Carbon (Pg C)	1946.1	69.0 (3.6)	183.5 (9.4)	351.9 (18.1)	−303.4 (−15.6)	−315.8 (−15.7)	−335.5 (−15.8)	−365.1 (−15.9)	627.7 (32.3)	649.5 (32.2)	689.9 (32.4)	758.4 (33.0)
Total Ecosystem Nitrogen (Pg N)	79.6	3.4 (4.3)	9.1 (11.5)	17.7 (22.2)	−8.9 (−11.2)	−9.4 (−11.3)	−10.1 (−11.4)	−11.3 (−11.6)	16.8 (21.1)	17.6 (21.2)	19.1 (21.5)	21.7 (22.3)

Table 2. Terrestrial Ecosystem Carbon (TEC) sensitivity to CO_2 fertilization (β_L) and its changes under increasing N deposition, TEC sensitivity to warming (γ_L) and its changes under increasing N deposition and TEC sensitivity to nitrogen deposition (δ_L) and its changes under increasing CO_2 concentration and warming. The pairs of experiments used to calculate the sensitivities are shown in Fig. 7.

	β_L (PgC ppm^{-1})	γ_L (PgC K^{-1})	δ_L (PgC (TgN yr^{-1})$^{-1}$)		
1N	2.21	−152	N deposition	With 2xCO2	With 2 K
2N	2.30	−158	3.41	4.48	2.79
4N	2.43	−167.7	3.01	4.03	2.48
8N	2.67	−182	2.47	3.39	2.04

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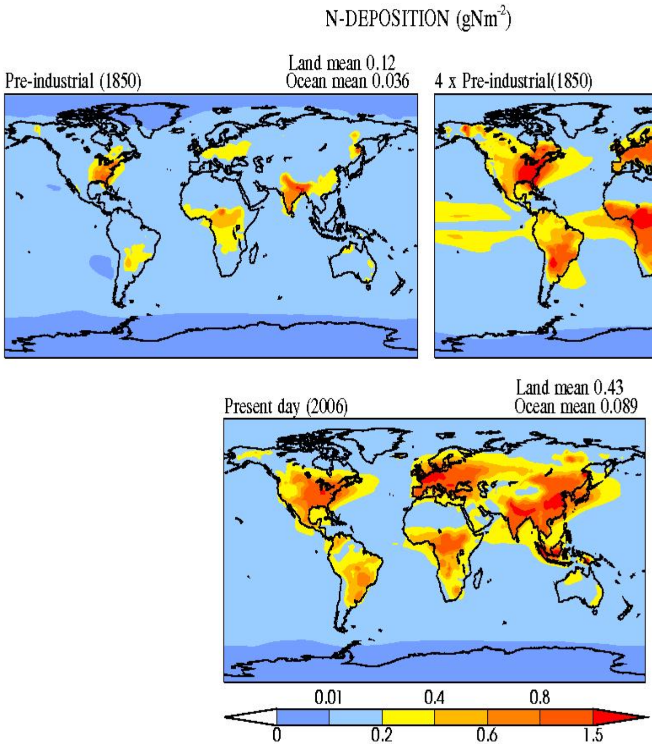

Fig. 1. Spatial distribution of N deposition in pre-industrial (1850; top left panel) period, four times the pre-industrial N deposition case (4N; top right panel) and present day (2006; bottom panel) in the input datasets of CLM4. The global mean pre-industrial N deposition over land is 0.12 gNm^{-2}(a total land deposition of 20.3 $TgNyr^{-1}$). The deposition in the experiment 4N (80.6 $TgNyr^{-1}$) is approximately close to the present day deposition of 0.43 gNm^{-2} ($\sim$ 73.1 $TgNyr^{-1}$). We infer from this figure that the spatial pattern of N deposition used in our experiments is similar to present day deposition.

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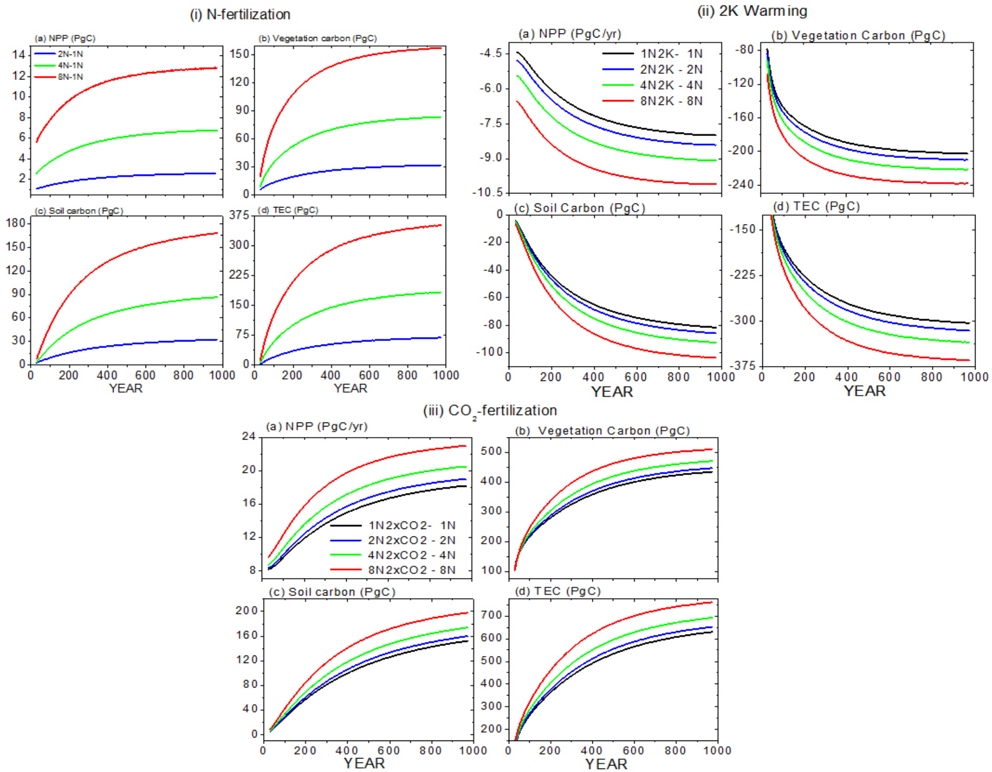

Fig. 2. CLM4 simulated global and annual mean changes in NPP, vegetation carbon, soil carbon and total ecosystem carbon for (i) N deposition, (ii) climate warming and (iii) CO_2-fertilization at various levels of N deposition. A 57 yr running average is applied to original annual mean data.

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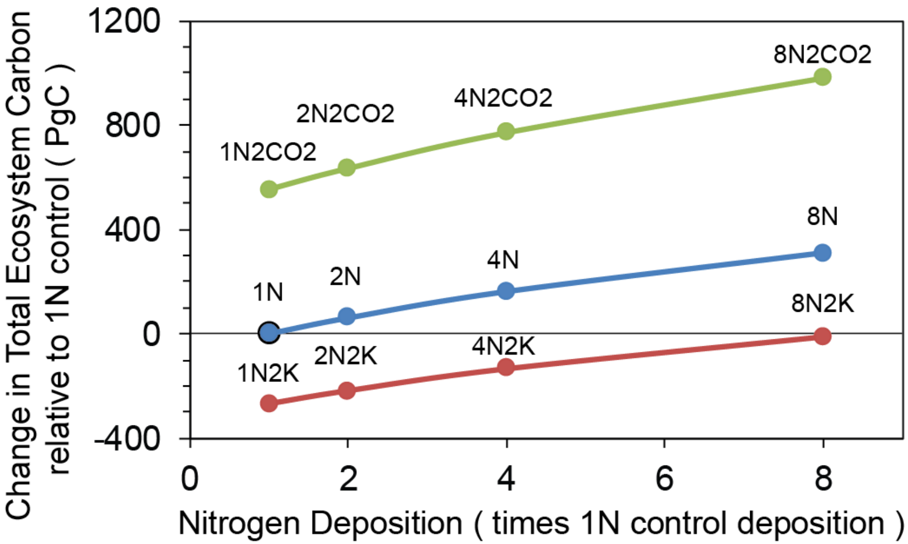

Fig. 3. Total Ecosystem Carbon (TEC) changes relative to the control simulation (1N) in the three sets of simulations. Blue line shows effect of increased N deposition. Green line shows effect of both doubled atmospheric CO_2 content and added N deposition. Brown line shows effect of both 2 K warming and added N deposition. The effect of an eight-fold increase in N deposition is approximately the same magnitude but opposite in sign to that of a 2 K warming.

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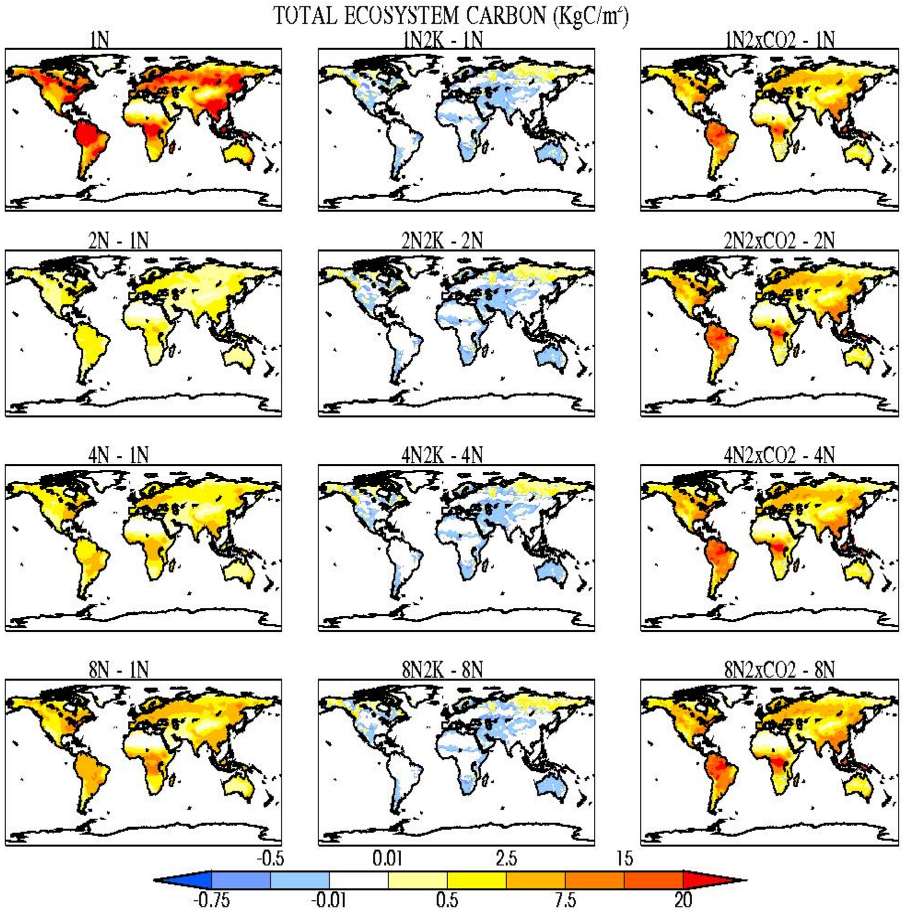

Fig. 4. Total Ecosystem Carbon (TEC) changes in the three sets of simulations. Left panels show TEC changes for N deposition and middle and right panels show TEC changes for climate warming and CO_2-fertilization under different background N deposition, respectively.

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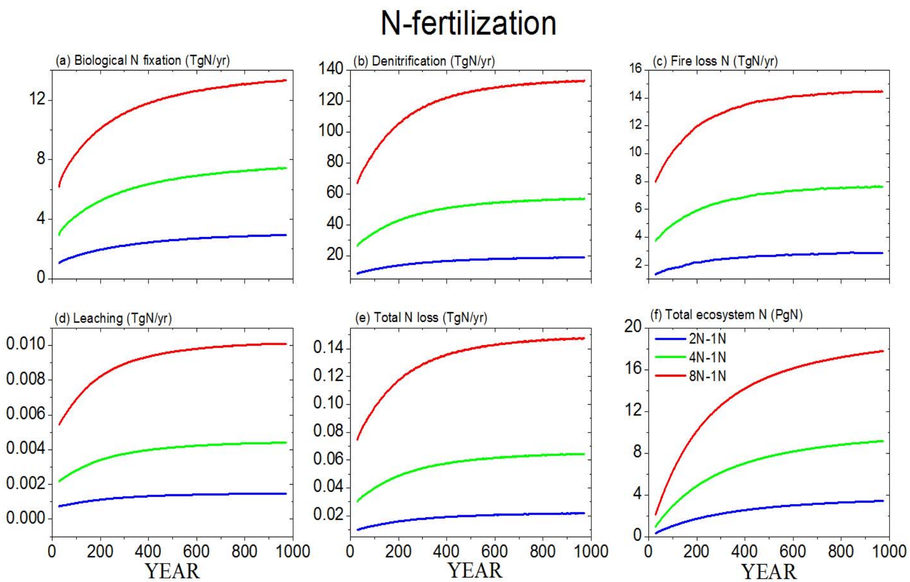

Fig. 5. Changes in N-budget for the terrestrial ecosystem in 2N (blue line), 4N (green line) and 8N (red line) simulations relative to 1N: changes in annual mean **(a)** biological N fixation (BNF), **(b)** denitrification, **(c)** fire loss N, **(d)** leaching, and **(e)** the total N loss from ecosystem (sum of denitrification, fire loss N and leaching). Total ecosystem N **(f)** is the cumulative sum of BNF and N deposition (constant in time for all simulations) minus cumulative sum of total N loss from ecosystem. A 57 yr running average is applied to original annual mean data.

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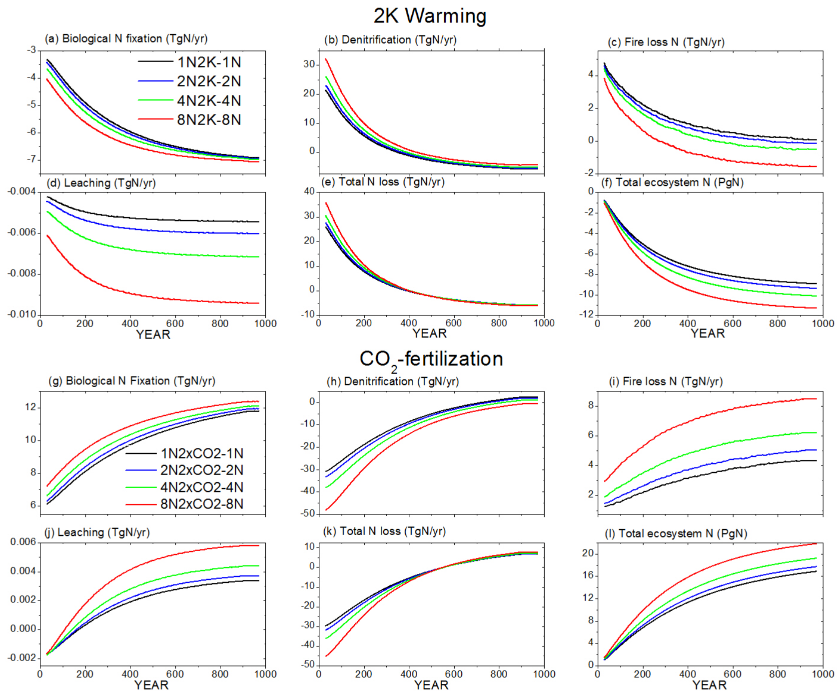

Fig. 6. Changes in N-variables for the terrestrial ecosystem in the 1000 yr simulations, 1N (black line), 2N (blue line), 4N (green line) and 8N (red line) in presence of 2 K warming **(a–f)** and CO_2 fertilization **(g–l)**: The global- and annual-mean changes of **(a, g)** biological N fixation (BNF), **(b, h)** denitrification, **(c, i)** fire loss N, **(d, j)** leaching, **(e, k)** total N loss from ecosystem (sum of denitrification, fire loss N and leaching) and **(f, l)** total ecosystem N (TEN). The order of magnitude of N-fluxes indicates that denitrification flux is the dominant process controlling N-stock changes. We find that the TEN losses in **(e)** are higher in presence of 2 K warming at higher N deposition levels due to larger decline in biological N fixation **(a)** and increase in denitrification **(b)**. In the case of CO_2 fertilization, TEN gains are larger at higher N deposition levels because of larger increase in BNF and decline in denitrification.

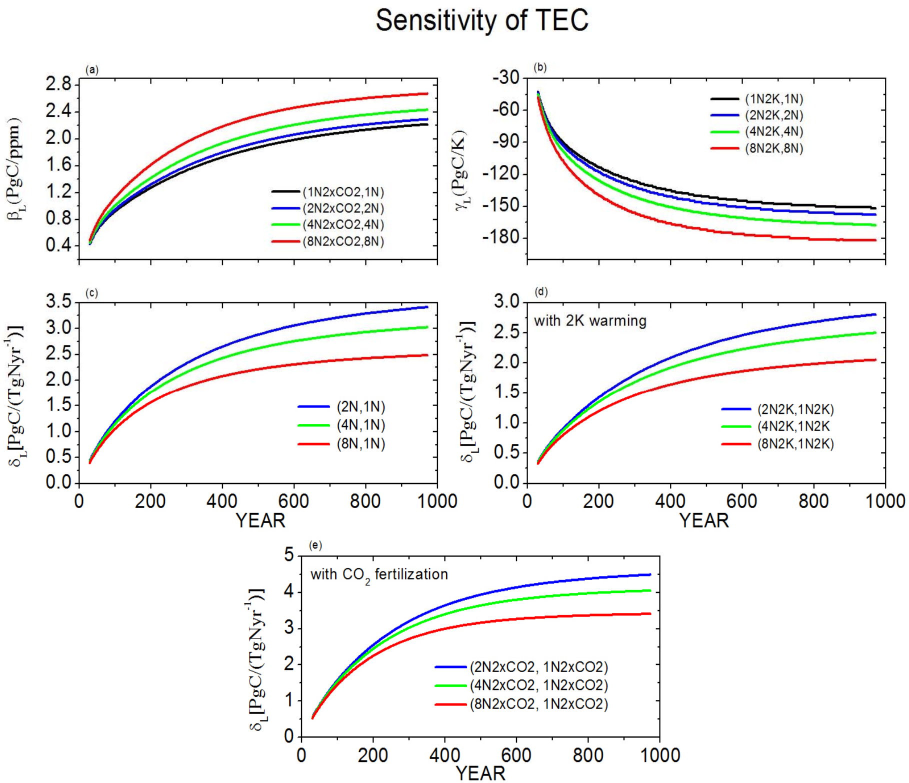

Fig. 7. Evolution of terrestrial ecosystem carbon (TEC) storage sensitivity to CO_2, climate warming and increased N deposition in our 1000 yr simulations. TEC sensitivity to **(a)** atmospheric CO_2(β_L) at different levels of N deposition, **(b)** temperature (γ_L) at different levels of N deposition, **(c)** N deposition (δ_L), **(d)** N deposition (δ_L) in presence of 2K warming and **(e)** N deposition (δ_L) in presence of doubled CO_2. A 57 yr running average is applied to original annual mean TEC data. The pairs of experiments indicated in the legend are the experiments that are used to calculate the respective sensitivities (Eq. 1–3).

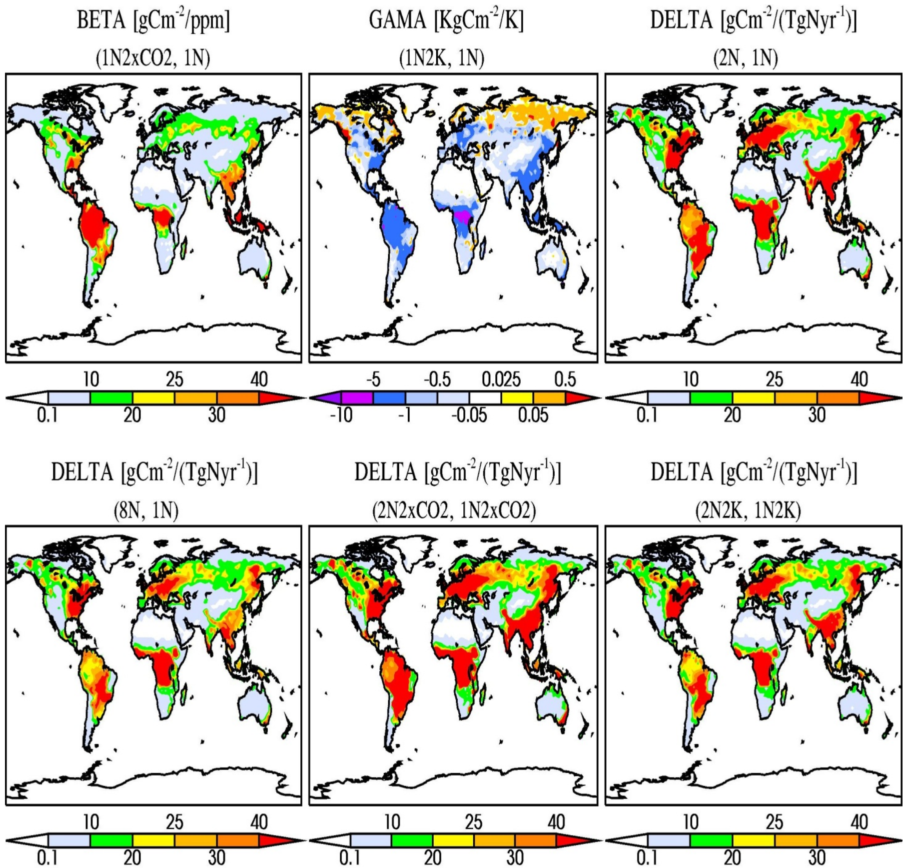

Fig. 8. Spatial pattern of total ecosystem carbon (TEC) sensitivity for CO_2 change (β_L), climate change (γ_L) and N deposition (δ_L; top panels). The experiments used in the calculation of sensitivity are shown in the parenthesis. The bottom panels illustrate the changes in delta for larger changes in N deposition (general decline), and in the presence of CO_2-fertilization (increase) and climate change (decline).

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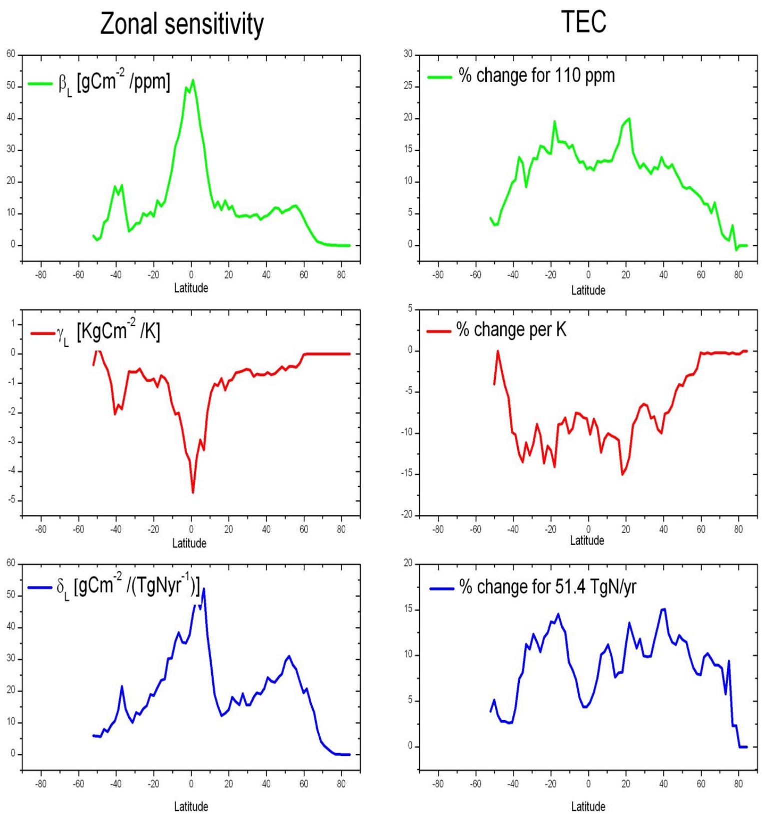

Fig. 9. Zonal mean pattern of total ecosystem carbon (TEC) sensitivity for CO_2 change (β_L), climate change (γ_L) and N deposition (δ_L; left panels). The right panels show the percentage changes in TEC during the historical period due to CO_2 change (110 ppm), climate warming (~ 1 K) and N deposition (51.4 Tg N yr^{-1}).