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Modelling the demand for new nitrogen fixation by terrestrial ecosystems

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Abstract. Continual input of reactive nitrogen (N) is required to support the natural turnover of N in terrestrial ecosystems. This "N demand" can be satisfied in various ways including biological N fixation (BNF) (the dominant pathway under natural conditions), lightning-induced abiotic N fixation, N uptake from sedimentary substrates, and N deposition from natural and anthropogenic sources. We estimated the global new N fixation demand (NNF), i.e. the total new N input required to sustain net primary production (NPP) in non-agricultural terrestrial ecosystems regardless of its origin, using a N-enabled global dynamic vegetation model (DyN-LPJ). DyN-LPJ does not explicitly simulate BNF; rather, it estimates total NNF using a mass balance criterion and assumes that this demand is met from one source or another. The model was run in steady state, and then in transient mode driven by recent changes in CO_2 concentration and climate. A range of values for key stoichiometric parameters was considered, based on recently published analyses. Modelled NPP, and C:N ratios of litter and soil organic matter, were consistent with independent estimates. Modelled geographic patterns of ecosystem NNF were similar to other analyses, but actual estimated values exceeded recent estimates of global BNF. The results were sensitive to a few key parameters: the fraction of litter carbon respired to CO_2 during decomposition, and plant type-specific C:N ratios of litter and soil. The modelled annual NNF increased by about 15% during the course of the transient run, mainly due to increasing CO_2 concentration. The model did not overestimate recent terrestrial carbon uptake, suggesting that the increase in NNF demand has so far been met. Rising CO_2 is further increasing the NNF demand, while the future capacity of N sources to support this is unknown.

1 Introduction

Terrestrial plant growth depends on net primary production (NPP), which is what remains of total photosynthetic carbon (C) fixation (gross primary production, GPP) after plant respiration has returned about half of the GPP to the atmospheric carbon dioxide (CO_2) pool. Global terrestrial NPP is about 50-60 Pg C a^{-1}. NPP is approximately balanced by the transfer of plant matter to detritus (litter), which is decomposed by microbial action to become soil organic matter (SOM)

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with the release of much of its C content as CO_2. Eventually the SOM itself is also oxidized to CO_2. In steady state, NPP must equal the total release of CO_2 from the decomposition of litter and SOM, plus a small contribution from fire. With rising atmospheric CO_2, rates of photosynthesis and NPP can increase and therefore C stocks can increase, allowing net uptake of anthropogenic CO_2 (Ciais et al., 2014). However, plant tissues contain elements in addition to carbon, hydrogen and oxygen – most abundantly nitrogen (N), which originates as N_2 in the atmosphere but must be supplied to plants in reactive forms including nitrate (NO_3^-) and ammonium (NH_4^+). N is repeatedly recycled between plants and soil: when inorganic N is released (mineralized) from litter and SOM during decomposition, it becomes available for re-uptake by plants (or microbes). A large fraction of the total N stock in most ecosystems is recycled in this way. But the cycle is not closed. N is lost through leaching (both dissolved and particulate forms are taken along with flows of water in the soil, and transferred to streams and rivers), and as gases: ammonia (NH_3) emitted by volatilization, and nitric oxide (NO), nitrous oxide (N_2O) and dinitrogen (N_2) emitted by microbial processes, principally denitrification. These losses have to be replenished by new supplies of reactive N for a steady NPP to be maintained, and the supply rate has to increase further if NPP and C storage are to increase. We refer to this requirement for new reactive N supplies from any source (not only BNF) as the 'new N fixation demand' (NNF) of terrestrial ecosystems.

There are large uncertainties in current knowledge of the N inputs to terrestrial ecosystems. Biological nitrogen fixation (BNF), and to a lesser extent nitrogen oxide (NO_x = NO + NO_2) production from N_2 by lightning, are the main natural processes that can satisfy the N demand of ecosystems. Early estimates of global terrestrial BNF were 90-130 Tg N a^{-1} (Galloway et al., 1995) and 100-290 Tg N a^{-1} (Cleveland et al., 1999), based on upscaling field measurements. But recent global estimates are much lower, e.g. 58 (40-100) Tg N a^{-1} (Vitousek et al., 2013). (Sullivan et al., 2014) suggested downgrading conventional estimates of BNF in tropical forests (generally regarded as a hotspot of N fixation) by a factor of five, based on new measurements. Early large estimates of the lightning contribution to N fixation (> 100 Tg N a^{-1}: (Liaw et al., 1990) have also been revised downwards, to 1-20 Tg N a^{-1} (Labrador, 2005). NO_x emissions from soils (and fires) can be transported in the atmosphere and subjected to dry or wet deposition in other places, but this is a small flux to terrestrial ecosystems: about 4.5 Tg N a^{-1} for oxidized N species (NO_y) and 13 Tg N a^{-1} for reduced species (NH_x) (Galloway et al., 1995). Human activities have altered the global N cycle through the widespread use of N fertilizer, whereby atmospheric N_2 is initially fixed by the Haber-Bosch process, and the release of reactive N to the atmosphere through fossil fuel burning. Global agricultural N inputs have been estimated as ~140 Tg N a^{-1} (Galloway et al., 1995;Schlesinger, 2009) and total N deposition over land as ~50 Tg N a^{-1} (Dentener et al., 2006). But the fate of most fertilizer N is to be either lost in gaseous emissions, or leached out of the fields and transported away in streams. Enhanced atmospheric N deposition is concentrated near populous industrialized regions, resulting in N saturation or even overload in some places, but with limited effect over most of the global land surface(Cleveland et al., 2013). BNF remains as the largest likely contributor to satisfying terrestrial ecosystems' new N demand in a global perspective, while uncertainty surrounds the actual magnitudes both of the global new N demand and of the extent to which it is satisfied by BNF. Moreover, rising CO_2 concentration and the resulting increase in GPP have inevitably further increased the new N demand . Thus three key knowledge gaps are (1) the magnitude

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of the global new N demand; (2) the magnitude of terrestrial BNF, and its ability to satisfy demand; and (3) to what extent, and by what mechanisms, terrestrial ecosystems have been able to respond to CO_2-induced increases in N demand through the enhanced acquisition of N.

Model-based analyses have not yet cast much light on these issues as there is still no consensus on how to represent the coupling of the terrestrial C and N cycles. The first dynamic global vegetation models (DGVMs) did not consider N cycle processes at all. (Hungate et al., 2003) first drew attention to the large discrepancy between early 'optimistic' DGVM projections of high rates of carbon uptake in a high-CO_2 world (Cramer et al., 2001) and independent projections of N uptake based on contemporary rates. This analysis set a value of ~ 90 Tg N a^{-1} for current terrestrial BNF (Galloway et al., 2002). Several recent DGVMs have included strong N supply limitations on both NPP and the response of NPP to increasing CO_2 concentration, yet the process most likely to limit NPP in the long term – that is, BNF – has been represented in indirect ways: for example, as a function of actual evapotranspiration (Yang et al., 2009;Zaehle and Friend, 2010), based on earlier analyses by (Schimel et al., 1996) and (Cleveland et al., 1999), or simply as a function of NPP (see the discussion by (Wieder et al., 2015)) Some models have prescribed rather than predicted BNF (Houlton et al., 2008;Gerber et al., 2010;Esser et al., 2011). The basis for modelling N inputs to ecosystems thus remains largely unresolved. In this paper, we use a mass-balance approach, as implemented in the DyN-LPJ model of Xu-Ri & Prentice (2008), to address the question: how much newly fixed N *must* be made available each year, globally, from any source, in order to sustain NPP ? In other words, what is the 'demand' for newly fixed N for terrestrial ecosystem – and how can it be satisfied, based on current understanding of supply-side constraints? The DyN-LPJ model of (Xu-Ri & Prentice, 2008), which has also been used to quantify the N_2O-climate feedback (Xu-Ri *et al.*, 2012; Stocker *et al.*, 2013), takes a different approach from other models. It assumes that annual N supply f must not only balance losses of N, but also provide sufficient new N inputs to maintain the observed stoichiometry of litter, decomposer biomass and SOM. The model calculates the demand for newly fixed N(NNF) that is created by the enrichment of soil organic matter in N during litter decomposition. This demand cannot be fully met by recycling (N uptake and immobilization) from the soil inorganic N pool. The mass-balance calculation needed to quantify this demand involves the C:N ratios of plant litter and soil organic matter and the fraction of litter C that is respired to CO_2. We make use of recently published analyses of observational and experimental data on these parameters to constrain the demand for fixed N, and we model transient changes in demand based on observed changes in CO_2 concentration and climate.

2 Materials and Methods

2.1 Model description

DyN-LPJ has been used previously to quantify the N_2O-climate feedback (Xu-Ri et al., 2012;Stocker et al., 2013). The model is based on the Lund-Potsdam-Jena (LPJ) DGVM framework (Sitch *et al.*, 2003). In addition to the coupled carbon and water cycling and vegetation dynamics processes simulated by LPJ, DyN simulates the flows of N through atmosphere,

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vegetation, litter and soil, and back into the atmosphere. It includes submodels for plant N uptake, N allocation, turnover, reproduction, and mortality, plant and soil N mineralization, BNF, nitrification, NH_3 volatilization, nitrate leaching, denitrification, and N_2, N_2O and NO production and emission.

Fig. 1 illustrates the modelled stocks and flows of N. The full dynamic N mass-balance equations are listed in Appendix S1. The structure of the model is as described by (Xu-Ri and Prentice, 2008), except for the addition of a key feature that is essential for this analysis: namely the representation of immobilization – the uptake of inorganic N into microbial biomass – as the 'first choice' source of N to fuel decomposition. (In the earlier version of DyN-LPJ the inorganic N requirement of microbial growth was met from new inputs, resulting in an unrealistically high rate of total new N input.) During decomposition, an increase in litter N (net immobilization) may take place before release of litter N (net mineralization) begins. Typically, net mineralization only occurs after litter N concentration increased to a critical value. The *initial* chemical composition of the litter determines the critical C:N ratio (R_{CR}) at which this shift takes place (Parton et al., 2007;Manzoni et al., 2008) according to an empirical formula derived from litter decomposition experiments (Manzoni et al., 2008):

$$r_{CR} = 0.45\, r_L^{0.76} \quad (1)$$

where r_{CR} and r_L are N:C ratios, $r_{CR} = 1/R_{CR}$ and $r_L = 1/R_L$ where R_L is the litter C:N ratio.

Equation (1) expresses two important functional properties of the decomposer community. First, the kinetics of decomposition are determined by the initial litter chemical composition and do not change as decomposition proceeds. Second, decomposers that can break down carbon-rich litter also have a high critical C:N ratio corresponding to a low carbon use efficiency, $e = R_B/R_{CR}$ where R_B is the C:N ratio of the decomposer biomass (Manzoni et al., 2008). R_B does not vary systematically along gradients of organic matter or litter C:N, and typically remains in the range of 5 to 15. The fraction of litter C returned to the atmosphere by respiration is $1 - e$.

2.2 Climate and CO_2 forcing

A steady-state and a transient model run were set up using identical parameter values, spin-up protocols and forcings to the simulations described by (Xu-Ri et al., 2012) except that the transient run was repeated and extended to 2009, substituting TS 3.10.1 climate data (http://www.cru.uea.ac.uk/cru/data/hrg/) from the Climatic Research Unit, and updated atmospheric CO_2 concentration data from (Keeling et al., 2009), for the input data sets used previously. The contributions of climate and CO_2 changes to the transient simulation were assessed as in (Xu-Ri et al., 2012) by performing an additional transient run with time-varying climate but constant CO_2 (296 ppm).

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2.3 Sensitivity and uncertainty analysis

We considered the effect of varying R_S in the steady-state simulation from 4/5 to 5/4 of our central estimates (Tables 1, 2), a range corresponding to that found in the literature. We also examined the effect of varying e in the transient simulation. Many models, including the previously published version of DyN-LPJ, have set e = 0.3 (Sitch et al., 2003). This value was derived from the DEMETER model (Foley, 1995) and appears to have originated from CENTURY (Parton et al., 1992). Recent experimental determinations have indicated lower values of e, for example 0.25 in tropical Amazonian forest (Chambers et al., 2001) and 0.20 in temperate beech forest (Ngao et al., 2005). Assuming R_B = 10, the default value used by (Manzoni et al., 2008), results in a global average e of 0.23. The global average value of R_B has been estimated as ~7.6 (Xu et al., 2013), so the true global average value of e may be even lower (~0.175). Accordingly, we performed alternative model runs with R_B = 7.6 (low), 8.6 (intermediate) and 10 (high). The corresponding e values are 0.175 (low), 0.2 (intermediate) and 0.23 (high).

3 Results

3.1 Steady-state NNF

Global NPP in the steady-state run was 50.8 (49.6-51.3) Pg C a^{-1}, within the generally accepted range (Cramer et al., 1999). Total global ecosystem NNF was 340 (230-470) Tg N a^{-1} (Table 1). The geographic distribution of modelled NNF (Fig. 2) shows maxima in tropical forests and savannas, with tropical ecosystems (30°S-30°N) contributing 67% and northern extratropical ecosystems 30% to the global total. Ranges by biome were 4-10 g N m^{-2} a^{-1} in tropical ecosystems, 2-4 g N m^{-2} a^{-1} in humid subtropical forests, mediterranean-type ecosystems, maritime humid forests and boreal forests, and < 2 g N m^{-2} a^{-1} in temperate grasslands, tundra and desert.

The calculated NNF is influenced by the fraction of litter carbon respired to CO_2 during decomposition and plant functional type (PFT)-specific C:N ratios of litter and soil. Litter C:N ratios in the model are mainly determined by the PFT-specific C:N ratios of production (R_P, Table 2). The simulated global average litter C:N ratio in the model was 48.9 (Table 1), indistinguishable from 49.9 ± 3 as given in a recent review (Yang and Luo, 2011). The global average estimate of R_{CR} (~ 43) is close to the value of 40 estimated by Parton *et al*. (2007) and Manzoni *et al*. (2008). The global average modelled soil C:N ratio was 15.8 (Table 1), higher than the estimate of 13.3 by Post *et al*. (1985) but close to the recent value of 16.4 (Xu *et al.*, 2013) and lower than the value of 18.5 given by (Yang and Luo, 2011).

Uncertainty analysis of the steady-state run (Tables 1, 2) confirmed our expectation that lower soil C:N ratios (R_S) would increase the modeled NNF while higher values would decrease it. The C:N ratios of litter (R_L), in contrast, depend strongly on R_P and vary little among the simulations. A change of R_L between 48 and 50 (larger than simulated) would only change the critical C:N ratio (R_{CR}) from 42 to 43.5 (from eq. 1). Variation in R_{CR} through a larger range from 40 to 43 (Parton

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et al., 2007) only results in a change in modelled NNF from 340 to 360 Tg N a^{-1}. This uncertainty range is much smaller than that arising from the uncertainty in R_S.

3.2 Changes in NNF in response to changes in CO_2 and climate

Global NPP increased from 42.6 to 52.0 Pg C a^{-1} during the transient simulation. Lower, central and upper estimates of NNF (obtained by setting *e* at 0.175, 0.2 and 0.23) yielded increases through the same period from 290 to 340 Tg N a^{-1}, 340 to 410 Tg N a^{-1}, and 400 to 470 Tg N a^{-1} respectively (Fig. 3a). The increase in NNF was 40 to 60 Tg N a^{-1} (Fig. 3b) depending on the chosen value of *e*. About 80% of this increase was directly caused by the rising CO_2 concentration (Fig. 3a). The rate of increase in modelled NNF amounted to 0.47 to 0.67 Tg N a^{-1} for each ppm increase in CO_2 (Fig. 4d). Altogether about 76% of this additional NNF came from tropical ecosystems and about 17% from the northern extratropics (Fig. 3b), with a spatial pattern highlighting modelled hotspots of "woody thickening" in temperate and tropical savannas and woodlands (Fig. 5). There was a strong correlation between modelled NNF and NPP, both in terms of spatial (R^2 = 0.85) and temporal (R^2 = 0.86) patterns (Fig. 4b, c). The slope of the relationship was 0.007 to 0.009 g N g^{-1} C.

3.3 N losses and denitrification

Denitrification accounted for 71% of total modelled N loss. The modelled global denitrification rate, and the total N loss from terrestrial ecosystems, were from 180 to 240 and 260 to 340 TgN yr^{-1} respectively (Fig. 3c, d). In the transient simulation, N loss and denitrification rates were positively correlated (R^2 = 0.94). Both were more sensitive to climate than to CO_2 concentration (Fig. 3c, d; see also (Xu-Ri et al., 2012). The additional fixed N taken up in response to increasing CO_2 concentration was mainly stored in organic forms (Fig. 6a-c): on average 52% in SOM, 30% in litter, and the remainder in plant biomass.

The global terrestrial denitrification rate can be very roughly constrained by global natural land N_2O emissions, given assumptions about the N_2 to N_2O ratio in gaseous losses of N. The modeled global N_2O emission from unfertilized land was previously estimated as 8.6 Tg N a^{-1} (with a range of 7.6 to 10.5 Tg N a^{-1}) (Xu-Ri *et al.*, 2012), constrained by 66 worldwide measurements of total annual N_2O emissions from natural ecosystems. Modeled N_2 to N_2O ratios varied between 25 and 50 (Xu-Ri *et al.*, 2012), as determined by the maximum rate of N_2O production from denitrification in (Xu-Ri & Prentice, 2008). These values fall within the broad range of 20 to 220 from direct measurements of both fluxes made with a state-of-the-art technique (Dannenmann *et al.*, 2008).

3.4 NNF compared to N recycling between plant and soil

The total rate of N recycling from inorganic to organic compartments – equal to N uptake (0.98-1.05 Pg N a^{-1}, Table 1) plus immobilization (0.15 Pg N a^{-1}) – was estimated as 1.13-1.20 Pg N a^{-1}. The reverse flux – equal to litter (0.95-0.99 Pg N a^{-1}) plus SOM (0.44 to 0.69 Pg N a^{-1}) mineralization – was estimated as 1.39-1.68 Pg N a^{-1}. The imbalance between these

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two fluxes (recycling and mineralization) represents NNF, which has to be met from outside the 'loop' formed by plants and soil (Fig. 1). About 30% of current NPP (Fig. 7) was estimated to be supported by new N supplies, with maximal values of this fraction in tropical savannas, temperate semi-arid ecosystems and high-latitude ecosystems. Steady-state immobilization was 147 to 151 Tg N a^{-1}, about 10% of the total N mineralization rate (1.39-1.68 Pg N a^{-1}), consistent with experimental results (Hadas et al., 1992).

4 Discussion

4.1 Comparison with previous estimates of BNF

If BNF is assumed to be the largest supplier of N to terrestrial ecosystems, it makes sense to compare our estimated terrestrial ecosystems N demand for new N fixation (NNF) with independent estimates of BNF. However, our central estimate of global terrestrial N demand (340 Tg N a^{-1}) exceeds the *upper bound* of 290 Tg N a^{-1} given by Cleveland *et al.* (1999) for global terrestrial BNF, and exceeds more recent estimates (e.g. 127.5 Tg N a^{-1}, Cleveland *et al.*, 2013; 58 Tg N a^{-1}, (Vitousek et al., 2013) by a large factor. Our biome-average model estimates of N demand (Table 1) are similar to upper bounds of BNF given by (Cleveland et al., 1999) (Fig. 4a) while the model estimates generally greater N demand on a site-by-site basis than the (Cleveland et al., 1999) BNF data indicate, especially in high latitudes (Table 2). Thus there is an important gap between our model calculations of the N demand in non-agricultural ecosystems, and most estimates of the supply of newly fixed N through BNF.

There could be several reasons for this disparity, which we cannot currently distinguish. On the one hand, our model formulation may overestimate the N demand. It would be useful to compare our formulation with alternative modelling approaches to the estimation of total N demand. On the other hand, there is considerable heterogeneity among different estimates of BNF; some agents of BNF may not have been sufficiently considered; and other routes of entry for N may possibly be important. Some recent N fixation measurements based on the ^{15}N dilution technique have indicated that N fixation in alpine and temperate grasslands could be as high as > 1 g N m^{-2} a^{-1}, comparable with our estimates of N demand for these ecosystems (Yang et al., 2011). One recent analysis of 99 canopy trees in tropical forest also indicated a high fixation rate of 8-20 g N m^{-2} a^{-1} (Wurzburger and Hedin, 2016), comparable with our estimates of N demand in tropical ecosystems(Table 3). Additional N inputs derived from the weathering of fixed N in sedimentary rocks (Morford et al., 2011) may contribute significantly to meeting ecosystem N demand on deep soils (Mckinley et al., 2009). (Stocker et al., 2016) noted the remarkable diversity of natural N sources and the poor state of quantification of most of them, indicating a need for new field research to attempt to close ecosystem N budgets, especially in tropical ecosystems.

4.2 The fraction of NPP supported by newly fixed N

(Cleveland et al., 2013) provided estimates of the fraction of terrestrial NPP that is supported by newly fixed N, noting that an anlogous concept of 'new production' is well established in biological oceanography. They used satellite data to

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derive NPP and a method based on published syntheses of field measurements to derive the fraction of NPP supported by symbiotic and asymbiotic N fixation and N deposition. They estimated a total recycled N flux of 1.05 Pg N a^{-1}, similar to our estimated range of 0.98-1.05 Pg N a^{-1} (Table 1). Our modelled fraction of NPP supported by new fixed N in tropical ecosystems is much higher than in temperate and boreal forests(Fig. 7), in broad agreement with (Cleveland et al., 2013). However we estimated a larger fraction of total global NPP to be dependent on new N inputs (~30%, as opposed to 11% in Cleveland et al., 2013) due to our larger estimate of global ecosystem N demand(NNF).

Resorption from senescent leaves is an important pathway of nutrient recycling in most terrestrial ecosystems. Because resorbed N remains in the plant N pool and is subequently re-allocated during bud formation and early leaf expansion, increased N availability in soil might result in decreased N resorption (Brant and Chen, 2015;Lu et al., 2013). (Cleveland et al., 2013) estimated that about 30% of plant N demand was met by resorption. However, the N resorption flux remains within the plant N pool, and therefore does not contribute to the satisfaction of NNFas we define it. The impact of assuming that 30% of plant N uptake is obtained from resorption is illustrated by the cyan numbers in Fig. 1, whereby the plant N uptake decreases, initial C:N ratio of litter and N immobilization increases but NNF) s unchanged.

4.3 Has rising N demand been met?

The 'residual land sink' – that is, the uptake of CO_2 by those land ecosystems that have not been losing carbon due to deforestation – is estimated to have been 2.6 ± 1.2 Pg C a^{-1} during both the 1990s and the 2000s (Ciais et al., 2014), based on top-down calculations that are independent of terrestrial models. With C:N ratios for terrestrial organic matter in the range of 30 to 70 (De Vries et al., 2008;Sutton et al., 2008) it follows that the terrestrial N store must have increased at about 40 to 90 Tg N a^{-1}. This is consistent with our model estimates of a C:N ratio in the range of 35 to 50 (Table 1) and an increased NNF by 40 to 60 Tg N a^{-1}, with the additional N stored mainly in organic pools. Ciais et al. (2014) also drew attention to the need for increased N inputs to match terrestrial carbon uptake while maintaining stoichiometric constraints.

The rates of carbon uptake by the land during the 1990s and 2000s were modelled (central estimates) by DyN-LPJ as 1.7 and 1.8 Pg C a^{-1} respectively. Thus, the model *underestimated* the residual land sink. The rate of increase in the modelled terrestrial demand for N amounted to 0.47 to 0.67 Tg N a^{-1} for each ppm increase in CO_2 (Fig. 4d). Presumably, this increasing demand for N has been met, or exceeded, at a global scale; otherwise the observed terrestrial C uptake could not have occurred. This conclusion admits the possibility of increasing N limitation on NPP in some ecosystems, such as boreal forests, but nonethless poses a question as to the origin of the additional fixed N required to support carbon uptake on land.

4.4 N limitation and anthropogenic influences

It has been hypothesized that BNF might increase by 10-45% with CO_2 doubling (Hungate et al., 2003), but some experiments have suggested that increasing plant growth might not be sustained over many years of CO_2 elevation (Hungate et al., 2004) due to limitation of BNF and/or plant biomass accumulation by supplies of other elements. Strong N limitation of NPP has been reported in temperate and boreal forests (De Vries et al., 2006) and even in tropical forests (LeBauer and

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Treseder, 2008), while limited N supply has been mentioned frequently as a constraint on the CO_2 fertilization effect and has recently been shown to be a strong constraint on biomass increase in ecosystems dominated by arbiscular mycorrhizal symbioses (Terrer et al., 2016). On the other hand, 'mysterious N sources' have been invoked to sustain the increased carbon uptake by forests under long-term CO_2 enrichment (Mckinley et al., 2009). To some extent, CO_2-driven increases in NPP as observed in Free Air Carbon dioxide Enrichment (FACE) experiments may have been supported by increased exploration of the soil and increased rates of total N mineralization from SOM (Drake et al., 2011). (Zaehle et al., 2014) noted that the key process by which plants can acquire additional N to support CO_2-enhanced growth under N-limited conditions, as shown in some FACE experiments, is enhanced 'mining' of N from SOM. They found this to be a neglected process in DGVMs, with some models succeeding in reproducing observed CO_2-enhanced growth but for the wrong reason, i.e. due to an unrealistic degree of flexibility in the C:N ratio of plant biomass. But SOM 'mining' is presumably a process that has a time limit as potential N supplies in SOM are finite, reflecting the accumulation of a fraction of the N acquired by the ecosystem over time.

One non-mysterious source of newly fixed N is anthropogenic N deposition, which may have a synergistic effect with CO_2 in promoting enhanced NPP in temperate forests (Lloyd, 1999). Modelled NNF increased by 13-17% (average 15%) with increasing CO_2 (Fig. 3b), composed of 22-34 Tg N a^{-1} in the tropics and 13-19 Tg N N a^{-1} in the northern extratropics. According to (Dentener, 2006), atmospheric N deposition over land during the 1990s amounted to 22.5 Tg N a^{-1} in the tropics and 27.5 Tg N a^{-1} in the northern extratropics. Anthropogenic N deposition is thus of a large enough magnitude to have contributed significantly to satisfying increased NNF. However, its geographic distribution is extremely patchy. Most tropical and many temperate forests are remote from the large anthropogenic sources. When we compare the N supply by atmospheric N deposition (Dentener, 2006) with the modelled increase in NNF (Fig. 5) in the regions of heaviest N deposition (Europe, North America, South and East Asia) it appears that there is already an overload of N, i.e. more N is deposited than can be stored by organic components, in these regions; while other regions remain N-limited (Fig. 8).

5 Concluding remarks

Many authors have drawn attention to the need for increased N inputs to match terrestrial carbon uptake while maintaining the stoichiometry of plant and microbial life. Rising CO_2 concentration continues to increase natural ecosystems' demand for N at a global scale. Over multi-millennial time scales, it appears that new N inputs can increase sufficiently to support large increases in land carbon storage driven by increasing atmospheric CO_2 concentration, as took place over the last glacial-interglacial transition (Prentice et al., 2011). But the rate at which such adaptation can take place is unknown. Given the discrepancy between our mass-balance calculations and recent estimates of the rate at which newly fixed N enters the land biosphere, and considerable uncertainties surrounding this quantity, our impression is that current understanding of the sources of fixed N is insufficient to allow reliable process-based modelling of these sources. This discrepancy cannot plausibly be accounted for by N deposition or mining of N from SOM. The extent to which the supply of newly fixed N can

increase in response to increasing N demand is likewise unclear, and this knowledge gap remains an important uncertainty in model projections of the global C cycle. To address it will requires consideration of both the assumptions and implications of alternative numerical schemes to predict N demand, and empirical research to better quantify the components of total ecosystem N budgets.

Author contributions

X-R developed the model, performed the model simulations and evaluations, and wrote successive drafts. ICP participated in the model development, analysis and writing.

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Table 1 Modeled global NNF in steady state, including the range due to uncertainty in the soil C:N ratio (steady-state runs with e = 0.175). NNF, ecosystem demand for newly fixed N; N_{immo}, N immobilization rate; N_{up}, N uptake rate; N_{min}, N mineralization rate; NPP, net primary production; R_P, C:N ratio of production; R_v, C:N ratio of vegetation; R_L, C:N ratio of litter; R_S, C:N ratio of soil organic matter; R_E, C:N ratio of ecosystems.

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Experiment	NNF ($Tg\ N\ a^{-1}$)	N_{immo} ($Tg\ N\ a^{-1}$)	N_{up} ($Pg\ N\ a^{-1}$)	N_{min} ($Pg\ N\ a^{-1}$)	NPP ($Pg\ C\ a^{-1}$)	R_P	R_v	R_L	R_S	R_E
1 × central estimate of R_S	337.3	150.2	1.025	1.54	50.78	49.50	187.9	48.90	15.82	42.04
4/5 × central estimate of R_S	471.6	150.6	1.050	1.68	51.26	48.80	182.4	48.50	12.99	35.35
5/4 × central estimate of R_S	227.6	147.8	0.983	1.39	49.63	50.49	183.4	49.29	19.65	50.82

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Table 2 Prescribed C:N ratios for plant production (R_P) and soil organic matter (R_S) (McGuire et al., 1992;Xu-Ri and Prentice, 2008)

PFT	R_P	R_S (central estimate)	R_S (4/5 × central estimate)	R_S (5/4 × central estimate)
Tropical Broad-leaved Evergreen	43.75	16.73	13.38	20.91
Tropical Broad-leaved Raingreen	32.66	8.31	6.65	10.39
Temperate Needle-leaved Evergreen	89.17	23.86	19.09	29.83
Temperate Broad-leaved Evergreen	90.63	25.78	20.62	32.23
Temperate Broad-leaved Summergreen	65.00	20.09	16.07	25.11
Boreal Needle-leaved Evergreen	52.38	29.70	23.76	37.13
Boreal Needle-leaved Summergreen	45.24	18.15	14.52	22.69
Temperate Herbaceous	54.29	9.77	7.82	12.21
Tropical Herbaceous	69.55	10.34	8.27	12.93

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Table 3 Site-by-site comparison of modeled NNF (steady-state run, 340 ppm CO_2, with e = 0.175) with biological N fixation data summarized in (Cleveland et al., 1999).

Vegetation types	Longitude	Latitude	Location	Simulated NNF (g N m^{-2} a^{-1})	Range of N fixation rates in (Cleveland et al., 1999) (g N m^{-2} a^{-1})
Moist tundra and alpine tundra					
	–145.5	65.5	Alaska	2.40	0.28 to 0.94
	–113.5	53.5	Canada	1.67	
	16.5	62.5	Sweden	1.20	
Average				1.76	0.94
Boreal forest and boreal woodland					
	19	65	Sweden	1.29	0.1 to 0.3
	11.5	64	Norway	0.96	
	26.5	63	Finland	1.13	
Average				1.13	0.196
Temperate coniferous forest, deciduous forest and mixed forest					
	–114	50	Rocky Mountains	1.94	0.1 to 16
	–89	51	Ontario, Canada	1.30	
	12	47.5	Austria	1.58	
	175	–41	New Zealand	3.15	
Average				1.99	2.658
Temperate savanna, temperate tall grassland and short grassland					
	–93	45.5	USA	1.42	0.1 to 1
	–96.5	37	Oklahoma, USA	2.86	
	–105	41	Colorado, USA	1.38	
Average				1.89	0.305

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Tropical savanna and wet savanna					
	28.5	–24.5	South Africa	2.66	0.07 to 3.45
	–6.5	7.5	Ivory coast	6.53	
	6.5	9	Nigeria	4.82	
Average				4.67	4.400
Arid shrublands					
	-113	41	Utah, USA	1.33	3 to 9.75
	-68	-34	Argentina	1.18	
	-100.5	30.5	Southwest USA	3.06	
Average				1.86	3.393
Tropical evergreen forest					
	146.5	–7.5	New Guinea	6.60	0.1 to 24.3
	–72.5	3.5	Colombia	6.58	
	80.5	8.5	Sri Lanka	6.66	
	–156	19.5	Hawaii	4.13	
Average				5.99	3.607
Tropical nonforested floodplain					
	–53	–9	Brazil	7.40	0.63 to 24.3
Average				7.40	5.38
Tropical deciduous forest and tropical woodland					
	–1	6	Kade, Ghana	6.92	0.75 to 1.76
	83	25.5	Chakia, India	4.33	
Average				5.62	3.393
Desert					
	–117.5	35	Mojave	2.38	1 to 10
	–111.5	29.5	Sonoran	1.55	
	–117	40	Great Basin	1.93	
	130	–20.5	Australia	2.16	
	22	–23	Kalahari	1.90	
Average				2.00	1.078

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Figure captions

Figure 1 Schematic of stocks flows of N in steady state, as modeled by DyN-LPJ.

Figure 2 Geographic distribution of the modeled terrestrial ecosystems demand for newly fixed N (NNF, g N m^{-2} a^{-1}).

Figure 3 Transient simulations during the 20th century, with e = 0.175 and changes in CO_2 and climate, or climate alone: **(a)** Demand for newly fixed N (NNF, Tg N a^{-1}) **(b)** Increase in NNF due to rising CO_2 (by latitude bands) **(c)** Total N loss **(d)** Denitrification rate.

Figure 4 Modelled demand for newly fixed N, with e = 0.175: **(a)** Comparison of biome-average estimates with *upper bound* values from Cleveland *et al*. (1999) **(b)** Spatial relationship of NNF with NPP **(c)** Temporal relationship of NNF with NPP during the 20th century **(d)** Relationship of increased in global NNF to atmospheric CO_2 concentration.

Figure 5 Geographic distribution of the increase in NNF due to rising CO_2 (g N m^{-2} a^{-1}) **.**

Figure 6 Transient simulations during the 20th century, with e = 0.175 and changes in CO_2 and climate, or climate alone: **(a)** Ecosystem N balance **(b)** Organic N pool **(c)** Inorganic N pool.

Figure 7 Geographic distribution of the percentage of NPP supported by newly fixed N.

Figure 8 Excess of atmospheric N deposition over NNF during the 1990s (g N m^{-2} a^{-1}). Positive values imply N overload, negative values N limitation. The block structure is due to the coarse resolution of the N deposition input.

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Appendix S1. Dynamic N balance equations in DyN-LPJ

(1) $dN_{plant}/dt = N_{up} - N_{litterfall}$

(2) $dN_{litter}/dt = N_{litterfall} + N_{immo} - N_{minL}$

(3) $dN_{soil\_organic}/dt = N_{demand} + (1-f_a)\, N_{minL} - N_{minS}$

(4) $dN_{soil\_inorganic}/dt = f_a N_{minL} + N_{minS} - N_{up} - N_{immo} - N_{los}$;

In steady state:

$N_{minL} = f_a N_{minL} + (1-f_a)\, N_{minL}$

N_{minL} is the gross mineralization from litter, $f_a N_{minL}$ is the fraction of N in decomposed litter entering the soil inorganic nitrogen pool, and $(1-f_a)\, N_{minL}$ is the fraction of N in decomposed litter entering the soil organic matter pool. N_{minS} is the gross mineralization from soil. NNF, is the ecosystem demand for newly fixed N.

$dN_{organic\_pool}/dt = dN_{plant}/dt + dN_{litter}/dt + dN_{soil\_organic}/dt$

$dN_{organic\_pool}/dt = 0$

Combining (1) to (3), we obtain:

(5) $NNF + N_{up} + N_{immo} - f_a\, N_{minL} - N_{minS} = 0$

(6) $N_{minL} = NPP\ /R_{CR}$

(7) $N_{minS} = NPP\ (1 - f_a)/R_S$

(8) $N_{up} = NPP\ /R_P$

(9) $N_{immo} = NPP\ (1/R_{CR} - 1/R_L)$

(10) $R_P \approx R_L$

Combining (5) to (10):

(11) $NNF = (f_a\, N_{minL} + N_{minS}) - N_{up} - N_{immo}$, or

(12) $NNF = NPP\ (1 - f_a)(1/R_S - 1/R_{CR})$

For transient conditions Eq. (12) can be written as:

(13) $NNF = NPP\ (1 - f_a)(1/R_S - 1/R_{CR}) + dN_{organic\_pool}/dt$

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Figure 1

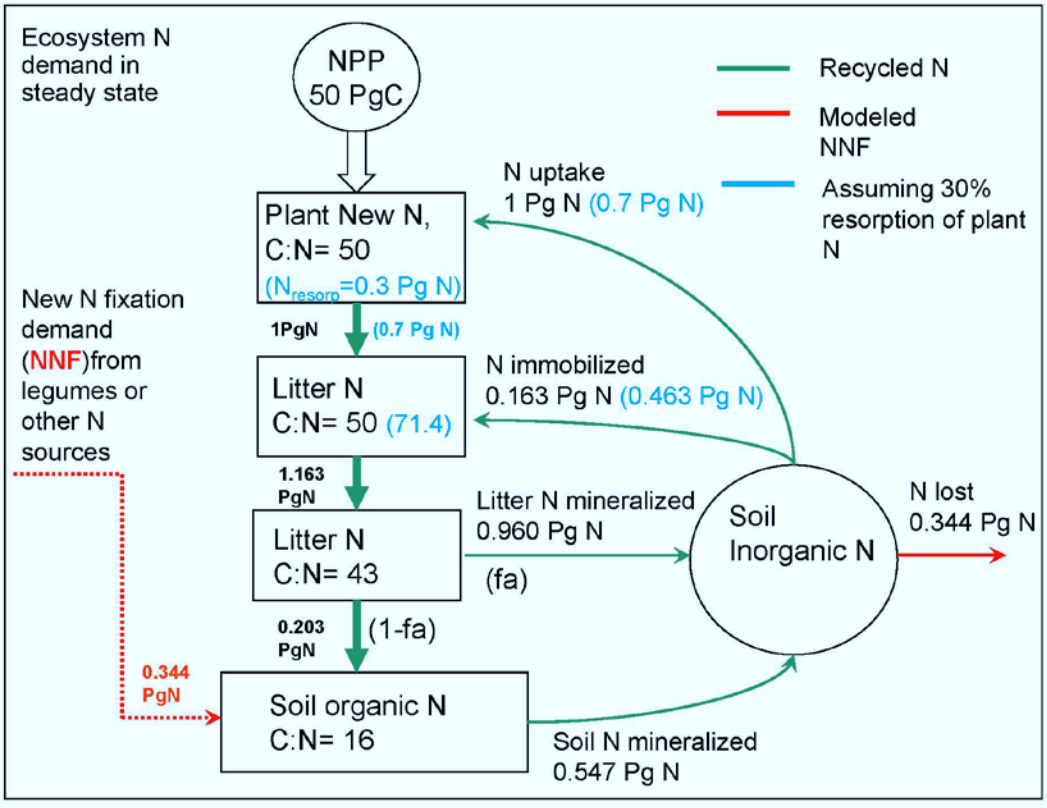

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Figure 2

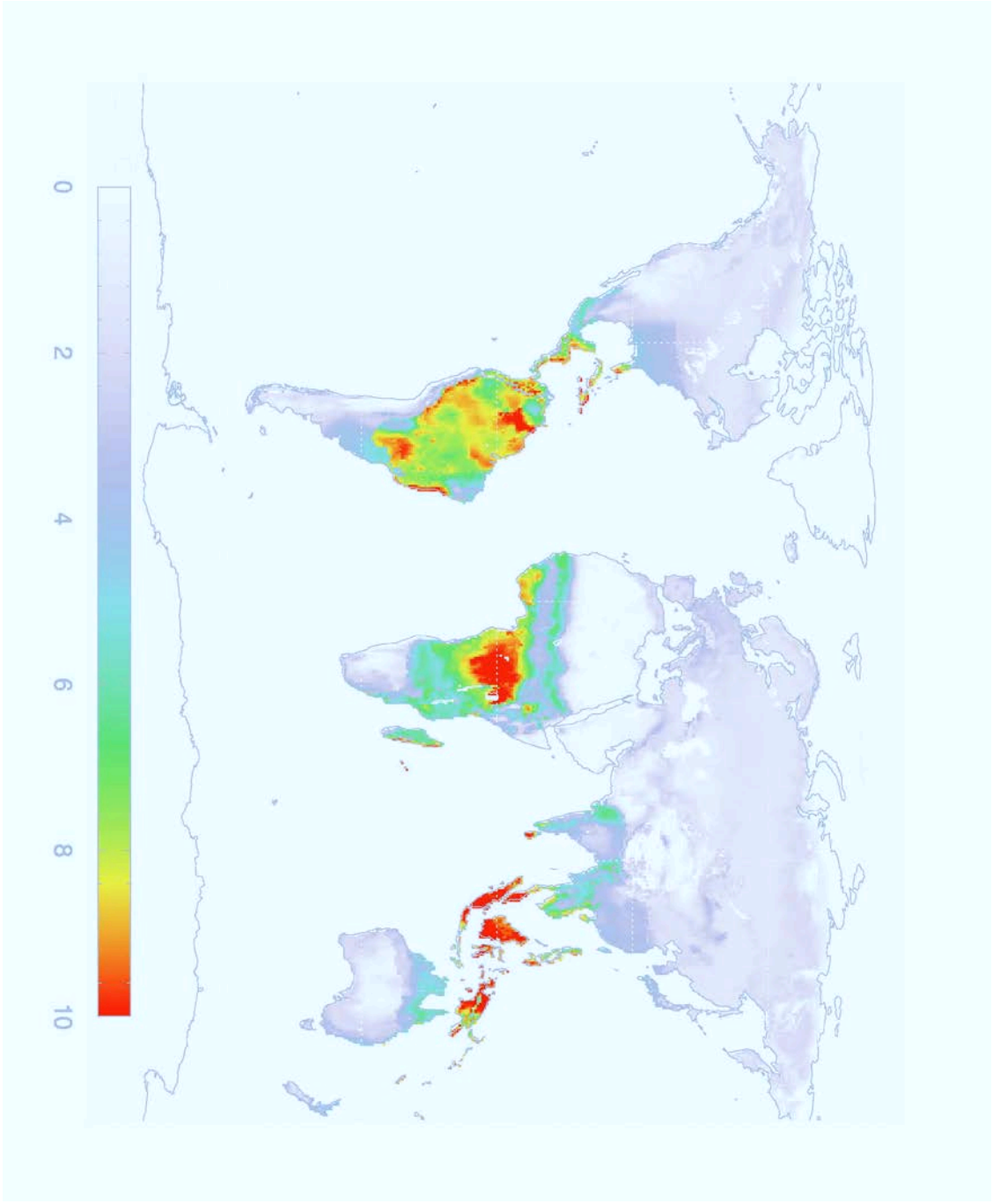

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Figure 3

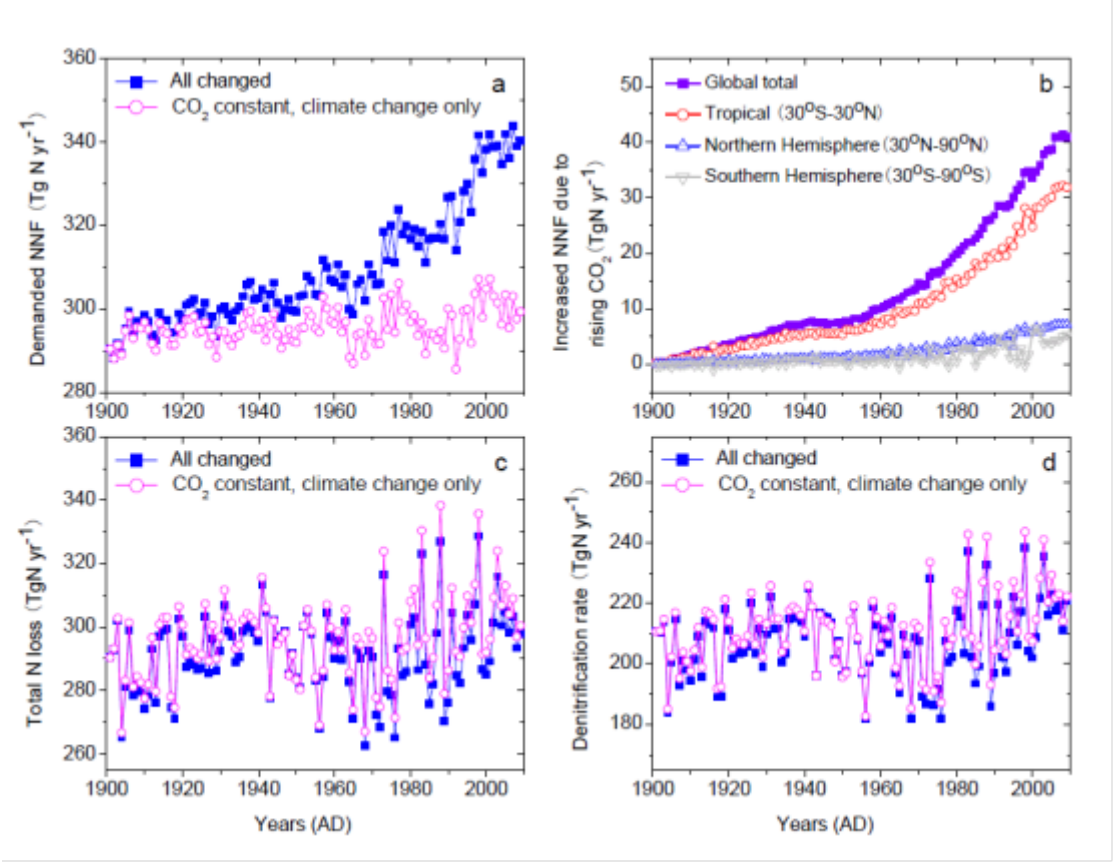

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Figure 4

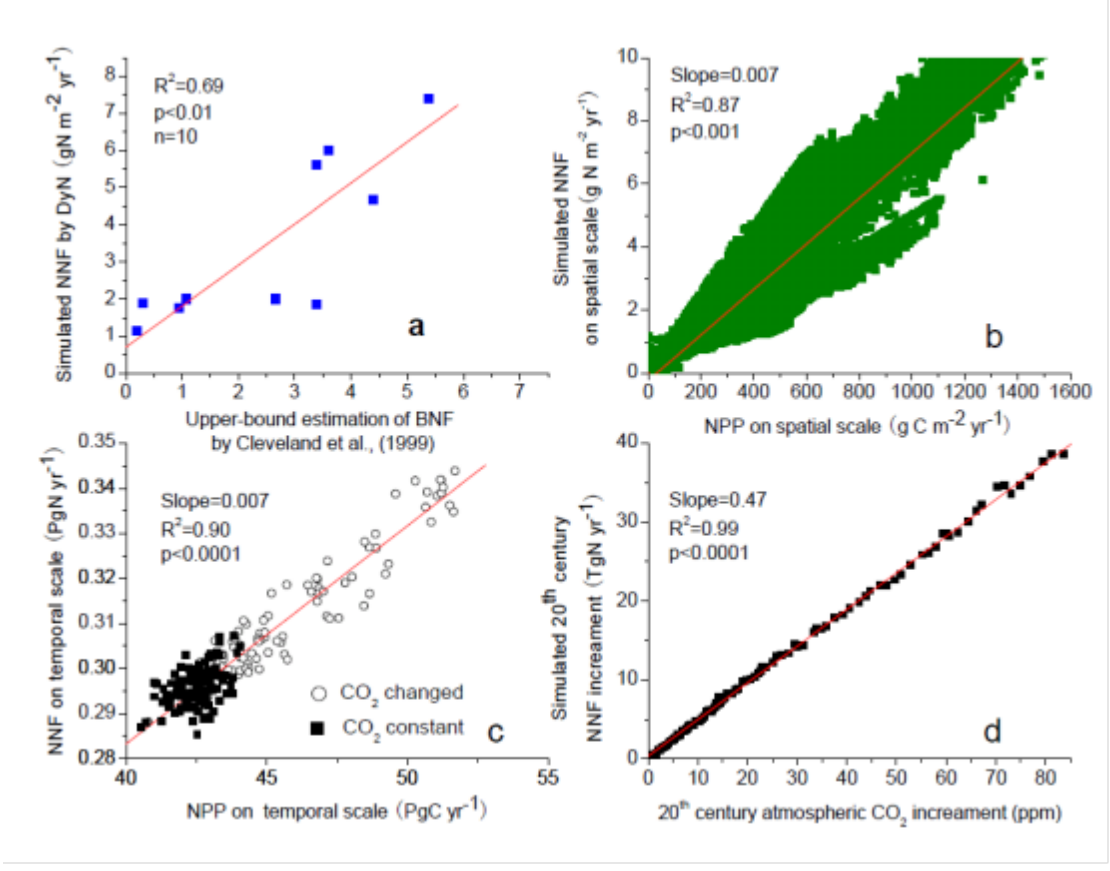

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Figure 5

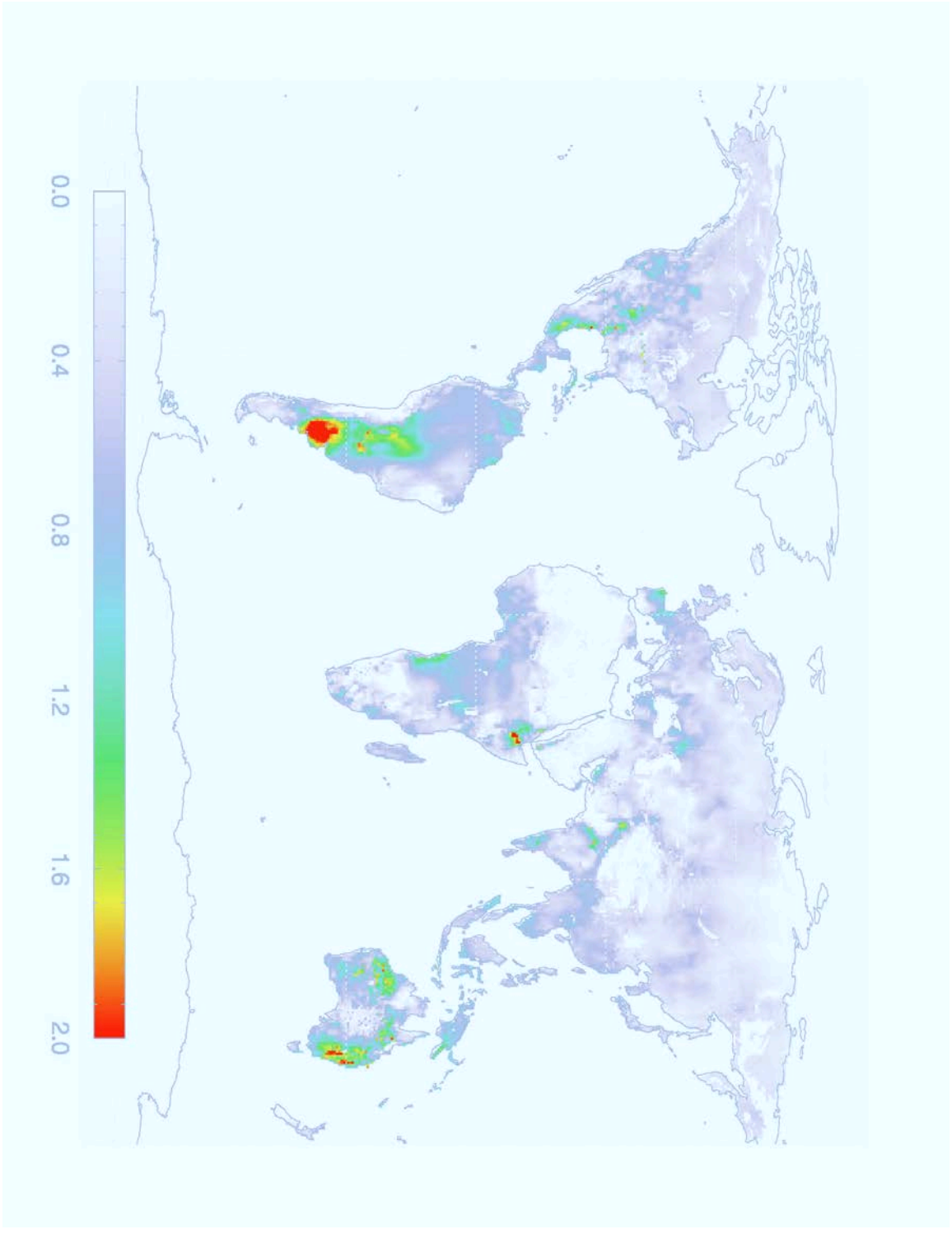

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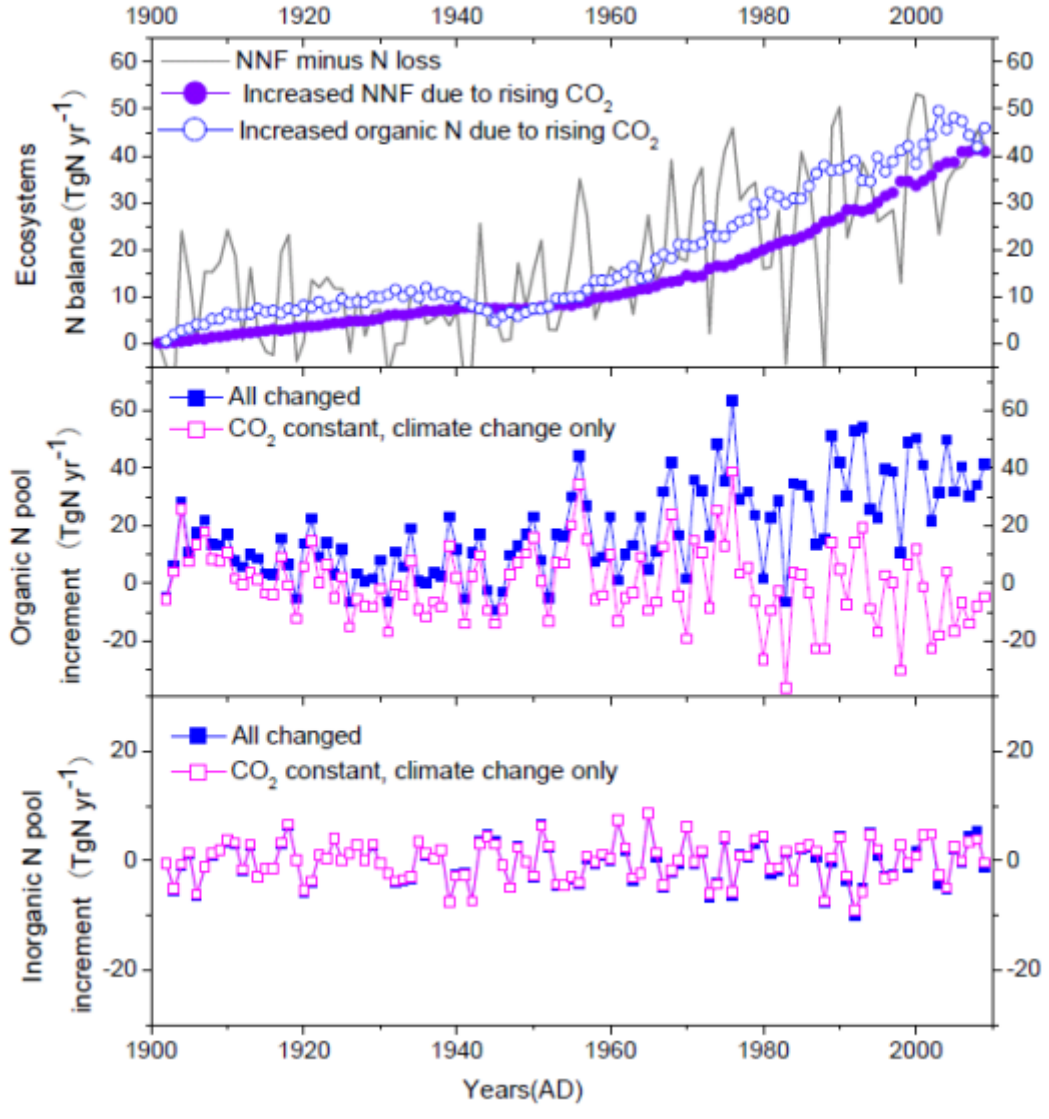
Figure 6



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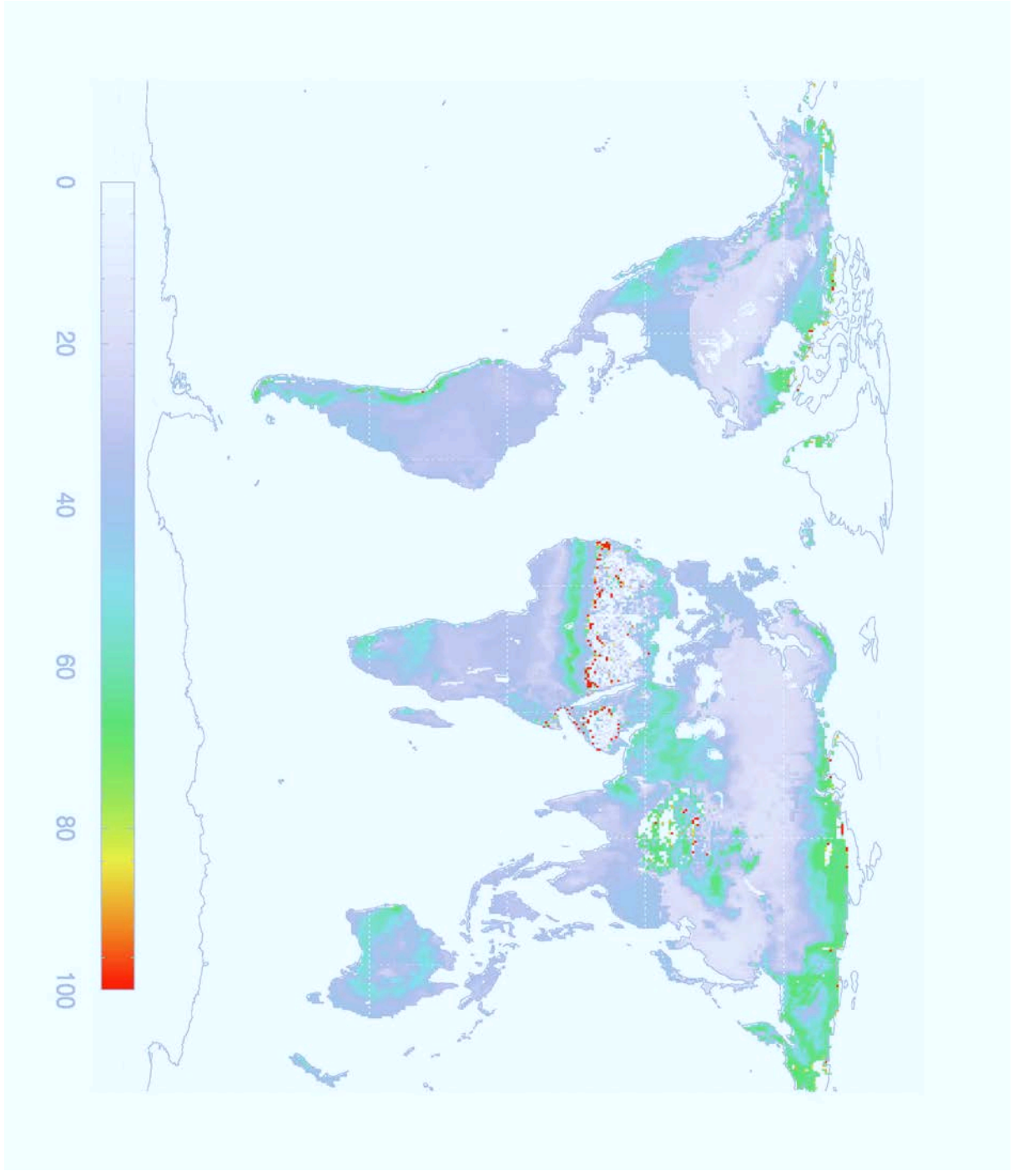

Figure 7

Biogeosciences Discuss., doi:10.5194/bg-2016-423, 2016
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Figure 8