Response to comments of reviewer 1

We thank the reviewer for the praise and the many helpful suggestions, which have largely been followed. Below our reply to the specific comments, set in red font color.

Braakhekke et al., use an Earth System Model to constrain the drivers of N leaching from 1901 to 2006. The manuscript is well written, interesting and contributes to a better understanding of the control of N leaching. Please find below a few comments that should be addressed before publication.

1) Atmospheric N deposition: theory and forcing

1.1. I am not really familiar with atmospheric N deposition, but from reading the manuscript it seems like the authors refer to nitrous oxide deposition, since N deposition is the highest in the most populated/polluted areas. I think a line or two properly defining atmospheric N deposition and its control could help better understand the paper.

Section 2.1.2 has been expanded with an explanation of N deposition and biological N fixation:

1.2. It is stated that the forcing for atmospheric N deposition is taken for years 1850-1860, whereas the other forcings (climate and CO2) are taken from year 1901. I guess there is not much difference between 1860 and 1901 for N deposition or at least much less than between 1901-2006, but a) where do estimates of N deposition for 1860 come from? And b) why would you take 1860 instead of 1901?

a) The N deposition data for the complete simulation period, including the spin-up, was obtained from the ACCMIP dataset Lamarque et al. (2013).

b) The aim of the spin-up is to obtain the ecosystem state under "pre-industrial" conditions, i.e. as little influence by humans as possible. For climate and CO_2 concentration anthropogenic effects are relatively small before 1900, compared to 1900-2000, but for N deposition regionally quite strong changes occurred before 1900 due to land-use change (biomass burning). Hence, we chose to use the first time period in the ACCMIP dataset, 1850–1860, both for the spin-up and the simulations where N deposition was held constant.

Section 2.2.1 has been modified to clarify the above two points.

1.3. Following on these 2 comments, I would suggest to restructure a bit section 3.1.1. as follow: The authors could start by describing Figure 1 and explaining the origin/controls of N deposition (natural vs anthropogenic effects, then describe Figure 2 and differences for each biome.

Done.

2) Comparison with previous estimates (Section 3.1.3) Beusen estimates seem to be significantly higher than LPJ-GUESS in Equatorial regions and southern tropics. The sentence L.13-15 is very unclear to me.

The relevant paragraph has been rewritten and is hopefully now more clear.

L. 4: "higher productivity in cold and dry regions at other latitudes." To me it seems more like in the "mid latitudes". East Australia and South Brazil/Argentina are not really cold-dry regions. I am not really

convinced by this section 3.1.3. I understand the authors want to try and compare their estimates with previous studies, but here a very rough comparison is made without going really into the reasons for these differences.

The line the reviewer referred to has been modified according to the reviewer's suggestion. An in-depth discussion on the reason of the mismatch for GPP is difficult without a detailed analysis including site observations and other data, which is outside of the scope of this study. Presumably, re-calibration would be needed to improve the fit. The aim of the comparisons with other data sets is to see whether LPJ-GUESS predictions are sufficiently realistic for the purpose of this is study, which—in our view—is the case, despite mismatch for GPP.

3) Climate section Due to the different impacts of temperature and precipitation changes on terrestrial productivity and soil processes, this section is a bit more difficult to follow. I would suggest the authors try to use terms which indicate the direction of the change: e.g. warmer conditions increase N mineralization...

We assume the reviewer is referring to section 4.1.2. The relevant paragraph has been slightly modified to meet the reviewer's request.

Due to the complicated relationship between climate and N leaching, as described in this section, it is not always clear what is the exact mechanism behind the predicted responses. Therefore, we prefer to formulate the discussion as we did: first mention the overall effect of climate change on N leaching and then discuss the likely reasons behind this response.

Minor comments:

- P2, L. 15: "with increasing N input, the capacity of ecosystems to retain N decreases.." that statement surprises me. I can understand that with increasing N input, leaching increases, but the capacity to retain N does not necessarily changes.
  "N input" has been changed to "N availability", which is more accurate.

- P4, L6. Add "be" between can and found.
  Done.

- P6, L. 27, "we"
  Done.

- P6, L.29: remove "to" between by and "the fraction"
  Done.

- Notation of "N leaching : N input ration", I don't; really like that notation as ":" also denotes a ratio. I would suggest to modify to "N leaching/N input" or "N leaching to N input ratio".
  All instances have been changed to "N leaching to N input ratio".

- P8, L. 20: add "is" after variability
  Done.

- Section 3.2.1, L. 18: N deposition and pCO2 increased but not climate, please modify.
  Done.

- P11, L.15, year of citation for Cleveland et al., is missing
  Since we already listed the publication year three lines previous, we deliberately omitted it in this sentence. The phrase has been changed from "the Cleveland et al. N fixation-AET relationship" to "the N fixation-AET relationship of Cleveland et al."

- Figures: not sure that showing the 99% quantile is the best way to go as areas with very large changes do not come out. You could also use a nonlinear color scale.
  The main reason to introduce the cutoff of the color axes in the maps are the extremely high N deposition and N leaching rates in Indonesia, as discussed in sections 3.1.1–3. These high values may not be realistic, and are of less relevance to this study since we are more interested in large-scale patterns. Since using a non-linear scale makes it more difficult to infer the absolute numbers from the graphs, we prefer to keep the color scales as they are.

- Figure 8 and 9: I find it a bit confusing that panel c does not go all the way to 90S. It would be better visually to have the latitudes of the 3 plots match: i.e. if c stops at 60S, then c does not have to be shown over the whole vertical plot and would stop at 60S in panels a and b).
  For all three panels in both figures (and the new figure), the lower limit of latitude is at 60°S.

Response to comments of reviewer 2

We thank the reviewer for the helpful comments which have led to an improvement of the paper. Below our reply to the specific comments, set in red font color.

Braakhekke et al. present a model study quantifying the respective contributions of changes in nitrogen (N) deposition, climate and atmospheric CO2 concentration on changes in N leaching from natural ecosystems. They find that increasing N deposition is the major driver behind simulated changes in N leaching, with smaller contribution from climate change and increasing CO2. They further highlight the role of fire in shaping N losses. The conclusions drawn have a sound basis on the the results discussed here. Overall, the manuscript is well written and clearly structured.

However, I find that the discussion of gaseous losses in the manuscript is lacking. Leaching losses are often the major loss, but gaseous losses are not negligible and can regionally dominated total N losses (Houlton et al. 2015). Accordingly, the role of gaseous losses needs to be considered when the ratio of leaching:inputs is discussed.

The author state in the introduction that "N leaching, while sometimes reported in global modelling studies, does generally not receive specific attention [...]". Therefore, due to the lack of evaluation of simulated loss terms, the reliability of global models in respect to the loss terms has to be considered low. This is true for LPJ as previous studies did not evaluate the loss fluxes sufficiently (Smith et al 2014 Warling et al., 2014). Unfortunately, the study by Braakhekke et al. does not improve this situation although data sets exist to evaluate. For example, the simulated gaseous loss fraction can be compared to reconstructions from delta 15 N measurements and models by Houlton et al. (2015) and, more recently Goll et al. (2017). There are regional differences in the dominant loss pathway between this study (Figure 3) and the mentioned studies which should be discussed.

We have added a figure and discussion on comparison of the fraction of N lost by denitrification to two global observation-based datasets: Wang et al. (2017) and Goll et al. (2017) (Figure 10). However, these two datasets differ considerably, demonstrating the current uncertainty regarding denitrification rates, also for observation based estimates.

The role of fire in shaping N loss pathways on global is a novel aspect of this study. The analysis would benefit from information on how simulated fire emissions and the contribution of wildfires to N deposition (forcing) compare to each other. Such information is completely lacking in the manuscript.

We acquired the dataset of N emissions from biomass burning used to derive N deposition. A figure has been added to the supplemental information (Figure S11), and the discussion on the role of fire (section 4.1.4) has been expanded.

In the abstract is stated "Predicted global N leaching from natural lands rose from 13.6 Tg N yr-1 in 1901–1911 to 18.5 Tg N yr-1 in 1997–2006, accounting for land-use changes." (P1L25/26). Did the authors account for land-use change? The information in the manuscript is insufficient to tell to what extent land use (change) and for example associated nitrogen fertilization was accounted for.

Since our study concerns only natural ecosystems, we did not consider N fertilization. However, to determine global total N leaching from natural lands, changes in natural land cover over time (mainly reduction) need to be considered. We did this simply by multiplying the fluxes by the natural landcover fraction for each grid cell. The sentence in the abstract to which the reviewer referred has been modified as to make this more clear: "*Predicted global N leaching from natural lands rose from 13.6 Tg N yr^{-1} in 1901–1911 to 18.5 Tg N yr^{-1} in 1997–2006, accounting for reductions of natural landc*over".

Minor P2L14: reference missing

Reference added.

P4L27: what is the criteria applied to define when the equilibrium state is reached?

During the spinup no checking is done to test how close the model is to equilibrium state. However, for soil organic carbon a root-finding solver is used midway in the spinup, to bring SOC pools very close to equilibrium. Testing has shown that this is sufficient.

P5L12: how are the grass PFTs being more competitive than trees in the model?

The higher competitiveness of grasses is achieved through PFT-specific parametrization, most importantly (cf also Smith et al., 2014):

1) Grasses have a higher uptake capacity per unit root biomass
2) In case insufficient N is available for all PFT (cohorts) N is partitioned among individuals according to a "relative uptake strength", which is higher for grasses.

P11:13: BNF estimates were revised down since Cleveland et al. 1999. Please account for newer estimates here; for example see Vitousek et al. 2013 , Sullivan et al. 2014.

We thank the reviewer for this good advice. The discussion has been updated to include the suggested references.

P14L23: The authors state that N deposition is the dominant factor driving spatial differences in the leaching rate. This needs to shown, as this is not apparent. I rather would suspect differences in the hydrological cycle to dominate spatial leaching patterns.

Our statement was largely based on the results of the factorial experiment (Figures 11 & 12, in the new manuscript). Because of the reviewer's comment we determined spatial covariation based on a moving window approach. This showed that both variation in precipitation and N deposition determine spatial patterns of N leaching, hence our statement was overly reductive. We removed it from the discussion.

P14L26: how is the correction done. This should be stated in the method sections.

This is described in section 2.3. We added a reference to this section.

Figure 4: the ratios, denitrification:inputs and fire:inputs, would be interesting to see and to better understand the lack of non-linearity in the simulated leaching:input ratio (Figure 6)

The suggested graphs have been added to the supplemental information (Fig S9 & S10).

P14L30: the substantial underestimation of BNF in LPJ should lead to lower leaching rates. This should be discussed.

A sentence has been added to the paragraph.

Reference: Houlton, Benjamin Z., Alison R. Marklein, and Edith Bai. "Representation of nitrogen in climate change forecasts." Nature Climate Change 5.5 (2015): 398-401.

Goll, D. S., Winkler, A. J., Raddatz, T., Dong, N., Prentice, I. C., Ciais, P., and Brovkin, V.: Carbon-nitrogen interactions in idealized simulations with JSBACH (version 3.10), Geosci. Model Dev. Discuss., doi:10.5194/gmd-2016-304, in review, 2017

Response to comments of reviewer 3

We thank the reviewer for the helpful comments. Below our reply set in red font color.

This paper presents a modeling study on global nitrogen leaching from natural ecosystems with an ecosystem model LPJ-GUESS. Overall, the paper is well-written and the results are informative. My major concern is on the dependence of results on model representation of nitrogen and carbon cycling process and the inherent model assumptions. Please find my specific comments below.

1. Authors conclude that atmospheric N deposition is the major driver behind nitrogen leaching globally. This is not surprising as atmospheric nitrogen deposition is the dominant N input and linearly linked to soil nitrogen storage in the model. I would suggest add more details on the mathematical formulations in representing atmospheric nitrogen deposition and nitrogen mineralization in the model. Are the results sensitive to the specific formulation of atmospheric nitrogen deposition and mineralization? Discussion on the nitrogen deposition dataset should also be added.

Nitrogen deposition was not predicted in this study, but taken from the dataset of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; Lamarque et al., 2013). This data comprises results from an ensemble of simulations with 11 complex atmospheric chemistry models. Therefore, providing details on the mathematical formulation of N deposition is not feasible. For this we refer to the Lamarque et al., (2013) and references therein. The results of the ACCMIP simulations have been thoroughly and favourably evaluated against observations by Lamarque et al. (2013), and can be assumed to represent the best estimate of global N deposition currently available. A sentence stating this has been added to section 2.2.1.

Regarding N mineralization, indeed the results are likely to be sensitive to the mathematical formulations in soil N cycling module. The soil N cycling module in LPJ-GUESS is largely based on the CENTURY model which has been applied many in many studies, and is described in detail in Smith et al. (2014) and Parton et al. (1993). A detailed discussion on the effect of different formulations of mineralization is outside the scope of this paper, also because we deem other aspects of the model more relevant for uncertainty of predicted N leaching (discussed in section 4.4). A thorough comparison of different N cycling models (including LPJ-GUESS) has been published by Zaehle et al. (2014), including discussions on the formulation of N mineralization.

2. Table 1 lists the numerical experiments conducted in this study. I would suggest add some statements on the purpose of these experiment designs in the methodology section 2.3. For example, which combination of experiments is used to disentangle the effects of a specific environmental driver (e.g. N deposition)?

Section 2.3 has been expanded with several sentences.

3. Increase in nitrogen deposition may potentially lead to increased plant carbon uptake and plant productivity, which would feedback to the nitrogen budget and affect leaching process. Is the impact of carbon–nitrogen interactions on N leaching process considered in this study? This aspect of carbon–nitrogen dynamics on N leaching process should at least be discussed. Please also add a table specifying

carbon–nitrogen ratios for all natural plant types considered. It would be interesting to examine/discuss how plant growth regulate the simulated N leaching for different PFTs in the model.

LPJ-GUESS is a dynamic global ecosystem model that simulates fully coupled C and N cycling in vegetation and soil. Hence, C-N interactions are considered and indeed an important aspect of this study (cf the effects of N deposition and CO2 on GPP, shown in supplemental figure S19).

Leaf C-N ratios are calculated prognostically based on canopy level photosynthesis and N availability (as described in supplement text S1), while C-N ratios of other pools are fixed. We added a figure of the PFT-mean leaf C:N ratios to the supplemental information (Figure S12)

4. The effects of fire and gaseous loss on N leaching is analyzed in the results section. But the descriptions on the representation of fire in the model is missing. In addition, it seems that the proposed numerical experiments in Table 1 doesn't consider fire?

Description of the fire module is given in supplemental text S1. This section has been expanded.

5. In section 2.2.1, the CRU monthly climate is interpolated to daily values as inputs for the model. More details on this temporal disaggregation are required. Discussions are also needed as the simulated sensitivity of N leaching to precipitation may depend on the daily sequence of precipitation and intensity.

Additional information on the interpolation of the climate data has been added to section 2.2.1. A sentence stating the relevance of rainfall distribution in time for N leaching has been added to section 4.1.2.

6. I would suggest clarify which specific aspect of N leaching is the focus of this study, the mean value or its temporal variation?

A sentence has been added to the introduction: "*Specifically, we focus on temporal changes during the last century in relation to change of environmental drivers, as well as spatial patterns of contemporary N leaching rates.*"

7. How is N status quantified?

This is done based on the N limitation factor, expressing reduction of photosynthesis due to N limitation. This is described in sections 2.1.2 and 2.3.

8. In section 3.2.1, the statement "N deposition, climate and atmospheric CO2 all increased during the 20th century" is confusing as "climate" is a broad concept.

The sentence has been modified.

9. I would suggest using the percentage change (%) as the unit in Figure 10

We assume the reviewer suggests to plot the relative changes as (sim-control)/control. We attempted this but it does not result in a readable graph, because regions where N leaching is very low in the control the relative change takes very high values, often infinity or NaN.

10. The name of the model used in this study can be added in the title.

We prefer to leave the title as it is. The name of the model is mentioned in the abstract.

Nitrogen leaching from natural ecosystems under global change: a modelling study

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Abstract. ~~In order to~~To study global nitrogen (N) leaching from natural ecosystems under changing N deposition, climate, and atmospheric CO_2, we performed a factorial model experiment for the period 1901–2006 with the N-enabled global terrestrial ecosystem model LPJ-GUESS. In eight global ~~simulations~~simulations, we used either the true transient time series of N deposition, climate, and atmospheric CO_2 as input, or kept combinations of these drivers constant at initial values. The results show that N deposition is globally the strongest driver of simulated N leaching, individually causing an increase of 88 % by 1997–2006, relative to pre-industrial conditions. Climate change led globally to a 31 % increase in N leaching, but the size and direction of change varied among global regions: leaching generally increased in regions with high soil organic carbon storage or high initial N status, and decreased in regions with a positive trend in vegetation productivity or decreasing precipitation. Rising atmospheric CO_2 generally caused decreased N leaching (33 % globally), with strongest effects in regions with high productivity and N availability. All drivers combined resulted in a rise of N leaching by 73 % with strongest increases in Europe, eastern North America and South-East Asia, where N deposition rates are highest. Decreases in N leaching were predicted for the Amazon and Northern India. We further found that N loss by fire regionally is a large term in the N budget, associated lower N leaching, particularly in semi-arid biomes. Predicted global N leaching from natural lands rose from 13.6 Tg N yr^{-1} in 1901–1911 to 18.5 Tg N yr^{-1} in 1997–2006, accounting for reductions of natural landcover~~land use changes~~. Ecosystem N status (quantified as the reduction of vegetation productivity due to N limitation) shows a similar positive temporal trend but large spatial variability. Interestingly, this variability is more strongly related to vegetation type than N input. Similarly, the relationship between N status and (relative) N leaching is highly variable due to confounding factors such as soil water fluxes, fire occurrence, and growing season length. Nevertheless, our results suggest that regions with very high N deposition rates are approaching a state of N saturation.

1 Introduction

During the last century, availability of mineral nitrogen for ecosystems across the globe has risen dramatically, mainly due to production and application of fertilizers and increasing atmospheric deposition, caused by N emission from fossil fuel combustion and agriculture (Bouwman et al., 2013b; Galloway et al., 2004). This increased N availability is thought to enhance terrestrial productivity and carbon uptake by relieving N limitation of natural ecosystems (Zaehle and Dalmonech, 2011). Excessive soil N input may, however, lead to N export, mainly as nitrate (NO_3^-), to ground- and surface water by leaching and lateral runoff. This results in a range of negative impacts on the environment and human health, such as eutrophication of fresh water and coastal ecosystems, fish kills, and reduction of drinking water quality (Rabalais, 2002; Schlesinger, 2009). In regions with severe ground- and surface water pollution most N export originates from agricultural land (van Egmond et al., 2002); hence these systems have been the focus of studies quantifying N budgets (e.g. Velthof et al., 2009). From a global perspective, however, natural ecosystems are a considerable source of N input to the hydrological system (Beusen et al., 2016a; van Drecht et al., 2003). In many ecosystems the combined input from biological N fixation and atmospheric deposition now exceeds the plant and microbial demand, and in some cases rivals fertilizer application in croplands (Dise et al., 2009).

With increasing N ~~input~~availability, the capacity of ecosystems to retain N decreases, resulting in larger leaching losses. However, the relationship between N inputs and mineral N leaching (hereafter simply "N leaching") is complex and non-linear, depending on factors such as vegetation type, climate, and soil properties. Insights from N manipulation experiments and measurements along N deposition gradients have spawned the concept of "N saturation", a state where N availability exceeds plant and soil microbial demand (Aber et al., 1989; Ågren and Bosatta, 1988). Since temperate forests have seen the largest increases in N deposition, previous work on N saturation and leaching has largely focused on these ecosystems. However, N deposition is spreading to regions that were previously less affected, including boreal, tropical, and (semi-) arid ecosystems (Galloway et al., 2004; Lamarque et al., 2013a). Response of N leaching in these ecosystems is likely to differ from that in temperate forests. For example, in many tropical forests N is not a limiting nutrient, due to high rates of biological N fixation and the limited phosphorous availability (Vitousek and Howarth, 1991). These ecosystems may thus be naturally close to N saturation (Matson et al., 2002) and N leaching is likely to be more responsive to changes in deposition (Matson et al., 1999). Grasslands, on the other hand, are usually N limited, but tend to occur in drier regions, where N losses are generally dominated by gaseous soil emissions (Bai et al., 2012) and emissions due to fire (Butterbach-Bahl et al., 2011).

Development of future N leaching is further influenced by global changes that affect terrestrial ecosystems, most importantly rising atmospheric CO_2 and temperature. Increasing CO_2 concentrations generally stimulate vegetation productivity (Norby and Zak, 2011), leading to increased N uptake (Finzi et al., 2007) and decreased N leaching (de Graaff et al., 2006; Hagedorn et al., 2000). The effects of higher temperatures on N leaching are more ambiguous: several warming experiments found a positive effect due to increased N mineralization (Beier et al., 2008; Rustad et al., 2001); but also absence of a

response (Beier et al., 2008) or even a negative effect due to increased vegetation activity (Patil et al., 2010) has been observed.

The combined effect of (changes in) drivers as well as ecosystem and soil properties results in complex spatial and temporal patterns of N leaching rates. Global prognostic models can help understand these patterns since they provide a means of upscaling process understanding from observational and experimental studies in order to assess N-cycling at large spatial scale. Many modelling studies on N leaching have been presented in the past decades, for both agricultural and natural ecosystems (e.g. Aber et al., 1997; Groenendijk et al., 2005; Li et al., 2006). However, ~~the majority of~~most of these models are computationally intensive and require site specific calibration; hence they are difficult to apply at global scale. Global prognostic models have been presented by van Drecht et al. (2003) and recently by Beusen et al. (2016), which provide spatially explicit estimates of N export to the hydrological system by partitioning N budgets into various flows, including leaching. While informative, these approaches rely on an equilibrium representation of terrestrial ecosystems and thus cannot represent changes in ecosystem N storage and N status. In this ~~context~~context, global terrestrial ecosystem models that explicitly include N cycling represent a complementary alternative. While less suited for site level application, these models include the most important ecological processes and feedbacks that influence N leaching, and are thus useful for examining spatial and temporal patterns and sensitivity to environmental factors. The main motivation to develop coupled C-N models has been to represent the constraint of N limitation on vegetation productivity and land C uptake (Zaehle and Dalmonech, 2011). Evaluation and application has therefore focused on variables related to C cycling (e.g. primary productivity, N use efficiency) rather than N cycling. N leaching, while sometimes reported in global modelling studies, does generally not receive specific attention (Gerber et al., 2010; Jain et al., 2009; Smith et al., 2014; Zaehle et al., 2014).

This paper presents a global modelling study ~~into~~of N leaching from natural ecosystems. Specifically, we ~~,~~focus on temporal changes during the last century in relation ~~and its response~~to change of environmental drivers, as well as spatial patterns of contemporary N leaching rates. We aimed to answer the following questions: 1) what is the effect of environmental drivers, most importantly N deposition, climate, and atmospheric CO_2 concentration, on N leaching from natural ecosystems? And 2) what is the current N status of natural ecosystems? We used LPJ-GUESS, a dynamic vegetation/ecosystem model optimized for regional and global studies that simulates terrestrial vegetation dynamics and biogeochemical cycles. The model has recently been extended to represent plant and soil N cycling and N limitations on plant productivity and carbon fluxes (Smith et al., 2014). The N-enabled version has been tested based on both site-level and regional observations (Smith et al., 2014; Wårlind et al., 2014) and includes the main processes underlying large scale patterns and global trends at decadal to centennial time scale of N leaching in response to drivers, which is the focus of this study. We present results from a global historical simulation, focusing on natural vegetation for the period 1901–2006. Predicted vegetation productivity and N leaching are compared to previously published estimates from measurements and models. Furthermore, to study the individual and combined effects of the main drivers of N leaching—N deposition, climate, and atmospheric CO_2—we

performed a full factorial experiment in which either the true transient time series were used for these drivers, or a trend-free time series, representative for pre-industrial conditions. We discuss the effects of these factors on N leaching in the context of insights from field observations, manipulation experiments and other modelling studies.

2   Methods

2.1   LPJ-GUESS

Here a brief overview of the LPJ-GUESS model is provided, focusing on processes that are most relevant for N cycling. A complete description of the model can be found in Smith et al. (2014) and references therein, as well as supplemental text S1.

2.1.1   General description

LPJ-GUESS (Lund-Potsdam-Jena General Ecosystem Simulator; Smith et al., 2001) simulates vegetation dynamics and biogeochemical fluxes of C and N in terrestrial ecosystems and employs generalized biome- or global-scale parameterizations of component ecosystem processes, allowing it to be employed without recalibration globally or for any large region without recalibration. LPJ-GUESS has been used extensively for studies from site to global scales. It is forced by climate variables, CO_2 concentration, and N deposition and runs with a daily time step, except for C allocation, vegetation dynamics, and disturbances, which are resolved annually. Our simulations focused on natural vegetation, i.e. croplands were not considered. Eleven plant functional types (PFTs) were included, representing vegetation in temperate, tropical and, boreal wooded ecosystems and grasslands. The model predicts the occurrence of each PFT based on bioclimatic limits and competition with other PFTs for light and soil resources. Contrary to most global ecosystem models, LPJ-GUESS explicitly represents the age distribution dynamics (demography) of woody PFTs and variations in stand development across landscapes, shown to be important for carbon and nutrient balance (Haverd et al., 2014; Wolf et al., 2011). The model simulates trees of different cohorts (age classes) of co-occurring PFTs, which are each represented by an average individual for each age class of each of a number of co-occurring PFTs. Mortality and establishment of the individuals are implemented in a stochastic fashion, as are fire (modeledmodelled according to Thonicke et al. (2001)) and other disturbances. Sub-grid variability resulting from landscape heterogeneity and differences in disturbance history are accounted for by simulating a predefined number of replicate "patches" (area 0.1 ha) per grid cell. The conditions for all patches within a grid cell are identical but differences arise from the stochastic calculations. Within each patch LPJ-GUESS simulates fluxes of C, water and N in vegetation and soil, based on descriptions of the key controlling processes, including photosynthesis, plant C allocation, autotrophic respiration, evapotranspiration, percolation, lateral runoff, and soil carbon cycling. The soil hydrological calculations are described in more detail in Gerten et al. (2004) and Olin et al. (2015). The simulation is

initialized with a ~~500 year~~500-year spin-up to accumulate vegetation and soil C and N pools in equilibrium with the initial forcing. During this ~~phase~~phase, the model is forced by a trend-free time series (here 10 years) of annually-varying inputs.

2.1.2 N cycling module

In LPJ-GUESS ecosystem N is present in vegetation biomass and in the soil in mineral and organic form. In the model
5   version employed for our study, mineral soil N is represented by a single pool; i.e. different N species such as ammonium and nitrate, and transformation between these are not distinguished.

~~In LPJ-GUESS ecosystem N is present in vegetation biomass and in the soil in mineral and organic form. In the model version employed for our study, mineral soil N is represented by a single pool; i.e. different N species such as ammonium~~
10   ~~and nitrate, and transformation between these are not distinguished.~~ Input of N in natural ecosystems occurs by biological N fixation and N deposition. Biological N fixation (BNF) refers to the uptake of atmospheric N_2, and reduction to ammonia, by free-living soil microorganisms and by symbiotic associations of microorganisms with plants. In LPJ-GUESS, BNF is calculated as a linear function of ecosystem evapotranspiration, following an empirical large-scale relationship identified by Cleveland et al. (1999). N deposition constitutes the input of reactive N from the atmosphere to the biosphere as particles
15   (dry deposition) or with precipitation (wet deposition). N deposition occurs as a range of species, both oxidized and reduced, and is controlled by N emissions from fossil fuel combustion, agricultural activities, and biomass burning—both natural and human induced. Since the residence time of reactive N species are relatively short (1-10 days), N deposition is a localized issue, and local rates are usually closely related to local emissions. N deposition rates are part of the model input (see section 2.2).

20   ~~Atmospheric~~N deposition and BNF ~~is~~are directly added to the soil mineral N pool~~, as is biological N fixation (BNF), which is calculated as a linear function of evapotranspiration following an empirical large-scale relationship identified by~~. Root uptake transfers N from the soil mineral N pool to vegetation on a daily time step. Plants take up N from the mineral soil pool in order to maintain optimal leaf N content required for photosynthesis (~~modeled~~modelled according to Haxeltine and Prentice, (1996)). Following Meyerholt and Zaehle, (2015), C ~~to :~~:N ratio of non-leaf biomass pools is fixed. If insufficient N
25   is available, the plant experiences N stress and photosynthesis is reduced. To this end the model calculates an "N limitation factor" equal to the ratio of the true V_{max} and the V_{max} in absence of N limitation (both without water limitation). Here, V_{max} is the carboxylation capacity of Rubisco. Additionally, different PFT cohorts compete for uptake of soil N, with grass PFTs being more competitive than tree PFTs.

N stored in vegetation is returned to the soil in organic form in conjunction with biomass turnover due to senescence,
30   mortality, and disturbance. Litter and soil organic matter (SOM) dynamics follow the CENTURY model (Parton et al.,

1993). Gaseous N loss during nitrification and denitrification is accounted for by a 1 % reduction of the daily N mineralization. Organic N leaching occurs as a fraction of the soil microbial N pool, determined by the percolation rate and the soil sand fraction. Mineral N leaching is calculated as a fraction of the mineral N pool equal to the relative water loss by percolation and interflow. N loss due to surface runoff is not considered. Finally, fire events cause loss of vegetation N, assumed to be emitted in gaseous form.

2.2 Global simulations

2.2.1 Forcing

The model was run globally on a 0.5° × 0.5° global grid. Climate forcing (mean monthly fields of temperature, precipitation, cloud fraction, and number of rain days per month) was taken from the Climate Research Unit (CRU) TS 3.0 data set (Mitchell and Jones, 2005; supplemental Figure S1b-c, S2, S3), and was interpolated to daily values. For precipitation, this was done using a stochastic weather generator, which randomly distributes monthly precipitation over the rain days in each month. For other variables, linear interpolation was used. Atmospheric CO_2 concentration was input as global means, varying annually (supplemental Figure S1d). Spatio-temporalal fields (interpolated to 0.5° × 0.5° resolution) of atmospheric N deposition were taken from the ACCMIP historical dataset (Lamarque et al., 2013; Figure 1Figure 1, S1a), which provides annual cycles with -monthly time steps for in decadal intervals for the period 1850–2010. This data comprises results from an ensemble of simulations with 11 atmospheric chemistry models, and can be assumed to represent the best estimate of global N deposition currently available. Ecosystem N input by deposition was not adjusted for leaf morphology.

During the spin-up, the model was run with forcing representing preindustrial conditions: forced by climate data for 1901–1910, detrended and cycled repeatedly, and, mean atmospheric N deposition for 1850–1860 and atmospheric CO_2 for 1901 (296 ppmv). Since N deposition has regionally undergone substantial changes before 1900 due to land-use change related biomass burning, we used N deposition for the first time step of the ACCMIP dataset, 1850–1860, both for the spin-up and the factorial experiment (section 2.3).

2.3 Factorial experiment

To disentangle the effects of N deposition, climate, and atmospheric CO_2 concentration on N leaching, we compared results for conducted a series of simulations in which the model was forced either by the true, transient values for these drivers, or trend-free time series as during the spin-up. We performed a full factorial experiment for the three drivers, resulting in the eight simulations shown listed in Table 1. Note that for this purpose atmospheric CO_2 concentration is not considered a climate variable. Herein we shall refer to simulation +Ndep +clim +CO_2 as the "true historical simulation", and -Ndep -clim -CO_2 as the "control simulation". These runs were performed with 20 replicate patches (section 2.1.1); all others with 10 patches, to limit computation time.

Comparison of results from the simulations in which one factor (e.g. N deposition) is included, to results from the control simulation allows us to study the individual effect of this factor. Similarly, interactions between factors can be studied based on the simulations with two factors included (e.g. N deposition and CO_2 increase). These runs were performed with 20 replicate patches (section 2.1.1); all others with 10 patches, to limit computation time.

5   For the analysis of the results we stratified the results by biome. Grid cells were classified into 17 biomes, based on leaf area index of the PFTs and latitude according to the scheme presented in Smith et al. (2014), which is based on Hickler et al. (2006).

We assess ecosystem N limitation and saturation based on the N limitation factor (section 2.1.2). This quantity serves as an indicator of vegetation N status and ranges between zero, signifying null rubisco capacity due to N limitation, and one, 10   signifying optimal rubisco capacity (no reduction due to N limitation).

Since the focus of this study is on natural ecosystems, we scale up LPJ-GUESS results to the globe assuming a world with potential natural vegetation. However, when comparing our results to other published estimates we correct them for the fraction of non-natural land based on the land use dataset of Hurtt et al. (2011). Models outputs are multiplied by natural land fractions (types "natural" and "barren") on a global 0.5° × 0.5° grid.

15   **2.4   Data used for model evaluation**

To evaluate our LPJ-GUESS predictionsresults, we compared several predicted variables to previously published estimates. First, predicted gross primary productivity (GPP) was evaluated based on a data-driven global product from the FLUXCOM dataset (Jung et al., 2017; Tramontana et al., 2016). This product was derived by training machine learning (ML) models on eddy-covariance measurements from the FLUXNET dataset using meteorological measurements and satellite data as input 20   data, and subsequently running these models for a global spatio-temporal grid. Estimates were made with three ML algorithms and two flux-partitioning approaches (Tramontana et al., 2016), resulting in six global products. We compare our results to the mean GPP and use the spread over the different products as a measure of uncertainty. The ML models were trained for 18 landcover types (LCTs) individually which were combined using area fractions of these LCTs, which wre derived from MODIS satellite data. Since we consider natural vegetation only, we derived a modified GPP as the weighted 25   mean over natural landcover types, where the weights are given by to the fraction of each LCT in a gridcellgrid cell divided by summed fraction for all natural LCTs.

Second, N leaching predictions were compared to estimates from the IMAGE model published by Beusen et al. (2016). Briefly, Beusen et al. modelled N flows using an equilibrium approach which partitions soil N input from deposition and fixation to various losses, including surface runoff, denitrification, and leaching. The model also included natural

ecosystems, but for the time frame of interest ~~gridcell~~grid cells were treated as either fully natural or fully anthropogenic. Therefore, we applied the same mask for natural lands to our predictions of N leaching to improve comparability.

Finally, the fraction of N lost in gaseous form by denitrification (f_{denit}) relative to total N loss (excluding fire) was compared to two observation-driven datasets derived by Wang et al. (2017) and Goll et al. (2017). These studies both used activity of the stable isotope 15-N in the soil to determine the denitrification fraction, based on a method published by Bai et al. (2012), but differ in their approach to upscale f_{denit} to the globe. Furthermore, since the isotope approach is mostly suitable for soils under natural vegetation (E. Bai, 2017 personal communication) Wang et al. used IMAGE model, to determine f_{denit} for croplands. Goll et al. did not treat croplands differently. Since it was not possible to correct the datasets for non-natural landcover, we compare the datasets to the LPJ-GUESS predictions as is. For more information on the observation based datasets, we refer to the respective publications.

3   Results

Presentation of the results focuses on the last ten years of the simulation (1997–2006), and their comparison to the pre-industrial baseline; most graphs (except time series) show model outputs for this period.

3.1   True historical simulation

3.1.1   Ecosystem N budget

Figure 1a and b depict N deposition, contemporary and changes relative to 1850–1860. Highest ~~rates of N~~ deposition rates ~~correspond~~occur in ~~to~~ human population ~~centers~~centres of Western Europe, Eastern USA, and South-East Asia (Figure 1~~Figure 1~~a). Since these regions are generally predicted to have temperate forests (supplemental Figure S4), these biomes have the highest N deposition and therefore overall N input (Figure 2~~Figure 2~~a, temperate broadleaf, deciduous and mixed forests). The same regions experienced the strongest increase in N deposition compared to pre-industrial conditions (Figure 1~~Figure 1~~b). Most of the world has experienced some increase in N deposition, with a few exceptions, most notably Florida, USA, where an assumed reduction in biomass burning in the N emission data set used to derive N deposition leads to reduced N deposition (van Aardenne et al., 2001; Lamarque et al., 2013a). Near the end of the 20th century Indonesia (particularly Kalimantan) shows extremely high N deposition with rates higher than 100 kg N ha⁻¹ yr⁻¹ (not apparent in Figure 1~~Figure 1~~ because the ~~color~~colour axis is cut off), a result of high N emissions caused by the severe forest fires in Indonesia in 1997 and 1998 (J.-F. Lamarque, personal communication, 2016).

Figure 2~~Figure 2~~ depicts the predicted ecosystem N budget for the 17 biomes and the world, including the simulated contribution of different export fluxes. Although within-biome variability is large, most biomes differ significantly from

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others for N input, N loss, and N net ecosystem exchange, as indicated by a Welch's t-test (supplemental tables S1–S3). Figure 2~~Figure 2~~b shows that all biomes have an average positive N net ecosystem exchange (NEE) over the 1997–2006 period, i.e. they are retaining N. ~~Highest rates of N deposition correspond to human population centers of Western Europe, Eastern USA, and South-East Asia (Figure 1a). Since these regions are generally predicted to have temperate forests (supplemental Figure S4), these biomes have the highest N deposition and therefore overall N input (Figure 2a, temperate broadleaf, deciduous and mixed forests). The same regions experienced the strongest increase in N deposition compared to pre-industrial conditions (Figure 1b). Most of the world has experienced some increase in N deposition, with a few exceptions, most notably Florida, USA, where an assumed reduction in biomass burning in the N emission data set used to derive N deposition leads to reduced N deposition (van Aardenne et al., 2001; Lamarque et al., 2013a). Near the end of the 20th century Indonesia (particularly Kalimantan) shows extremely high N deposition with rates higher than 100 kg N ha^{-1} yr^{-1} (not apparent in Figure 1 because the color axis is cut off), a result of high N emissions caused by the severe forest fires in Indonesia in 1997 and 1998 (J.-F. Lamarque, personal communication, 2016).~~ Biological N fixation (BNF; supplemental Figure S5) is less localized than deposition. Due to the empirical relationship to evapotranspiration assumed by the model of Cleveland et al. (1999), it is predicted to occur most strongly in the tropics, and is almost absent in deserts (Figure 2~~Figure 2~~a). However, in all biomes BNF is a less important N source than atmospheric deposition.

The relative contribution of N leaching, fire, and gaseous N loss to the total N loss varies strongly spatially (Figure 3~~Figure 3~~). N leaching (further discussed in section 3.1.2) is important in temperate regions and the tropics, mainly due to high inputs. N leaching also stands out in cold regions, particularly N. America, which is explained by temperature constraint on vegetation productivity. Conversely, in strongly arid regions (e.g. the Sahara) gaseous N emission dominates (c.f. also Figure 10a), due to low soil water fluxes. Finally, fire is an important N loss process in semi-arid regions, which mainly comprise grasslands. Predicted organic N leaching (supplemental Figure S6) is generally much lower than mineral N leaching, and is mostly negligible compared to the overall N budget.

3.1.2 Mineral N leaching and N status

Figure 4~~Figure 4~~a depicts N leaching for the true historical simulation. The regions where strongest N leaching rates occur generally correspond with regions of highest N deposition (Figure 1~~Figure 1~~). The American and African tropics show moderately high leaching, because of high BNF rates. The highest leaching rates (up to 95 kg N ha^{-1} yr^{-1}) occur in Indonesia due to the high N deposition rates for the target period (see section 3.1.1). The spatial patterns in Figure 4~~Figure 4~~a are largely mirrored by the N leaching to~~:~~ N input ratio (Figure 4~~Figure 4~~b), indicating that with higher N inputs the relative importance of leaching increases. Exceptions are regions with significant N losses due to mineralization (e.g. Russia, Canada), and cold regions.

Figure 5Figure 5 shows a global map of the N limitation factor, serving as an indicator of overall vegetation N status. It should be noted that in the model N limitation of photosynthesis is applied *before* moisture limitation (but *after* accounting for light and CO_2 concentration). Therefore, it is possible that in regions where both factors are limiting additional N input would not result in higher productivity. Biome dependencies are apparent (c.f. supplemental Figure S4) with grasslands and high latitude ecosystems having generally strong N limitation while temperate forests tend to be closer to N saturation. Deserts have a wide range of N status values. The Saharan and North American deserts are predicted to be strongly N limited. At high latitudes, the model also predicts very low N status for parts of the tundra and desert biomes. In most cases within-biome variability is partially explained by N input (supplemental Figure S8). However, the relationships differ strongly between biomes, and for the dry grassland and desert biomes it is virtually absent. Figure 5Figure 5 further shows that N status has risen during the previous century, owing mainly to increasing N deposition.

N status has a profound effect on the relationship between N input and N leaching per biome (Figure 6Figure 6). In strongly N limited biomes (tundra, grasslands, and boreal deciduous forest) considerably less N is lost by leaching than what is input. Semi-arid biomes (tropical deciduous forests, savannahs, and arid shrublands) also leach less N than they receive. However, this is mainly related to strong N losses due to fire (c.f. Figure 3Figure 3), which keeps these ecosystems continually in an aggrading state. The interaction between N status, fire, and N leaching is further illustrated by Figure 7Figure 7. The graph suggests a non-linear relationship between N status and relative leaching losses. However, this apparent non-linearity is mainly caused by biomes with high fire frequency which reduces the N leaching to N input ratio. An ancillary simulation run without fire disturbances resulted in a roughly linear relationship (supplemental Figure S14). Figure 7Figure 7 further confirms the exceptional position of deserts, where N cycling is mainly determined by physical processes rather than vegetation.

3.1.3   Comparison with previous estimates

Figure 8Figure 8 shows a comparison of gross primary productivity with estimates based on the FLUXCOM dataset (Jung et al., 2017). Although in general the spatial patterns are similar, LPJ-GUESS predicts lower productivity in the wet tropics and higher productivity in cold and dry regions at in mid other latitudes. These mismatches largely compensate each other, resulting in similar estimates for global total GPP (125.1 ± 6.9 PgC yr^{-1} (mean±SD) for FLUXCOM versus 116.8 PgC yr^{-1} for LPJ-GUESS). A similar result was found by Piao et al. (2013) for the C-only version of LPJ-GUESS.

The predicted mineral N leaching rates shows good agreement with estimates of the IMAGE model (Beusen et al., 2016; Figure 9Figure 9). The zonal mean by LPJ-GUESS shows a pronounced peak near the equator, which is not mirrored by N leaching from IMAGE results. This is likely explained by the high N deposition rates in Indonesia (section 3.1.1), which are not present in the N deposition map used as input for IMAGE (based on Dentener et al., 2006). Conversely, zonal mean N leaching is higher for IMAGE than for our results. The estimated gGlobal total N leaching from natural lands (adjusted for

changes in natural landcover) at near the end of the century by the two models compares similarly well, as shown in supplemental Figure S15b.l (Figure S12b). However, before 1990 LPJ-GUESS shows substantially lower global rates. Again, this is likely explained by differences in N deposition, which are higher for the Beusen et al. study, before 1990 (supplemental Figure S12S15a). Interestingly, the estimate of Beusen et al. is roughly constant throughout the 20th century (supplemental Figure S15b), which is explained by It appears that during the 20th century reduction of natural land and increases in N deposition approximately balance each other, resulting in roughly constant global total N leaching from natural ecosystems. a reduction of natural landcover, approximately balancing increases in N deposition per unit area of natural land. LPJ-GUESS predicts similarly roughly constant N leaching from natural lands before the 1970s, but increasing rates after that. A likely explanation for this difference between the two models is that LPJ-GUESS accounts for the effect of increasing temperature on N mineralization while the IMAGE estimate does not.

Figure 10 compares the fraction of N lost by denitrification relative to total N loss (f_{denit}) predicted by LPJ-GUESS to the two observation-based estimates of Wang et al. (2017) and Goll et al. (2017). Comparison of the maps reveals notable disagreement with regard to the spatial patterns of f_{denit}, also for the two observation-based datasets. Notwithstanding, all three estimates agree with respect to a tendency towards higher contribution of denitrification in dry regions (e.g. central North-America) and lower values in the wet tropics and regions with high N deposition. LPJ-GUESS does appear to underestimate the importance denitrification for the wet tropics, however, and over estimate it for northern latitudes in North America and Eurasia.

3.2   Factorial experiment

3.2.1   Changes in drivers during the simulation period

Global N deposition, climate and atmospheric CO_2 all increased changed substantially during the 20th century (supplemental Figure S1). Global total N deposition changed from 18.5 Tg N yr[-1] in 1850–1860 to more than 60 Tg N yr[-1] in 2000–2010, corresponding to a global mean N deposition of 1.4 kg N ha yr[-1] and 4.4 kg N ha[-1] yr[-1], respectively. Mean global land surface temperature rose by with more than 1° C (supplemental Figure S1b) with strongest changes in North America and Asia (supplemental Figure S2). Global mean precipitation (supplemental Figure S1c) does not show a strong trend, but regionally strong increases and decreases occurred (supplemental Figure S3). Atmospheric CO_2 concentration rose from 296 to 381 ppmv (supplemental Figure S1d).

3.2.2   Effects on N leaching

Figure 11Figure 10 depicts the N leaching difference relative to control for the single-factor simulations. N leaching for two-factor simulations is shown in supplemental Figure S147 (c.f. also supplemental Figure S136). For the simulation with true

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N deposition (Figure 11Figure 10a) N leaching response generally follows the change in N deposition, showing higher rates in most places except in Florida, USA, where N deposition showed a historical decrease in the forcing data (see section 3.1.1). Climate change on the other hand has varying effects (Figure 11Figure 10b). In regions with high organic carbon storage (Russia, Canada; supplemental Figure S103), or high N availability (Europe) the response of N leaching tends to be

5   positive, due to higher N release by mineralization. Several regions also show decreased leaching rates such as Northern India and Eastern Australia, which is mainly related to a reduction in precipitation. The effect of CO_2 increase (Figure 11Figure 10c) is generally negative, with strongest reductions in regions with high productivity, such as the tropics. From the three single-factor runs it is apparent that different drivers dominate in different regions for true historical simulation (Figure 11Figure 10d); e.g. rising N deposition in Europe and East Canada/USA, CO_2 increase in the western Amazon, and climate

10   (precipitation) change in Northern India.

Figure 12Figure 11 shows the global total N leaching for the eight simulations, assuming a world with potential natural vegetation only. The strongest single driver at the global scale is N deposition change, which by itself causes an increase of N leaching by 87 %. The overall effect of true climate is positive (31 % increase), indicating that the increased N mineralization outweighs possible uptake stimulation due to higher productivity. This is explained by the fact that the effects

15   of climate on global GPP are relatively small and mostly negative (supplemental Figures S158, S196). In contrast, the CO_2 effect on global GPP is strong, resulting in a pronounced negative effect on N leaching at the global scale (-33 %). Note that, the opposing effects of rising CO_2 and climate on N leaching roughly balance each other, particularly in the first half of the 20th century. Strong synergies between the drivers in relation to N leaching are not apparent from these results.

4   Discussion

20   ### 4.1   Controls on N leaching

4.1.1   N input

Figure 6Figure 6 shows that there are large differences between biomes with regard to the relationship between N input (deposition + BNF) and leaching. These differences are mainly related to N status: strongly N limited biomes have a lower slope, meaning that relatively less N will be leached out. Although very low N status is limited to a few biomes, in most grid

25   cells N limitation is relevant to some extent; only ~8 % of the grid cells have an N status of 0.95 or higher. However, ecosystems do not need to be fully N saturated before significant leaching is simulated. For many biomesbiomes, relative N loss by leaching starts to increase rapidly at around N status values of 0.6–0.7 (supplemental Figure S7) if water fluxes are high enough. Furthermore, increases in N deposition occur mostly in regions that historically already had high deposition rates (Wårlind et al., 2014). Hence, at the global scale the increase in N deposition results in a strong increase in N leaching.

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Several studies have reported an apparent threshold at 5–10 kg N ha^{-1} yr^{-1} in the relationship between N leaching and N deposition or throughfall in temperate forests. Below this deposition rate N leaching is constant at negligible levels, while above it leaching increases linearly with in deposition, although with high variability (Butterbach-Bahl et al., 2011; Dise et al., 2009). Approximately the same threshold was found for NO_3^- concentration in lakes and streams by Aber et al. (2003).

5   Furthermore, a recent study on the effect of N deposition on vegetation productivity concluded that photosynthetic capacity of forests reaches a plateau at approximately 8 kg N ha^{-1} yr^{-1} deposition (Fleischer et al., 2013). In the LPJ-GUESS predictions, similar non-linear response is observed for the arctic/alpine tundra and arid shrubland/steppe biomes (Figure 6Figure 6). For temperate forests, however, this behaviour is not apparent. Since the high within-biome variability may obscure the relationship, we plotted N deposition versus N leaching for the grid cells in Europe with temperate deciduous

10   forest together with data from Level II sites of UN-ECE/EC Intensive Monitoring Programme (supplemental Figure S2017; de Vries et al., 2003; Dise et al., 2009). This does not change the essential behaviour: there is in general no N input rate below which N leaching is negligible for all cells. However, the minimum N leaching (bottom of the cloud) shows an apparent threshold between 10 and 15 kg N ha^{-1} yr^{-1}. Below this rate cells with negligible leaching occur while above it virtually all cells show significant leaching.

15   Globally, biological N fixation rate (BNF) is predicted to be 18 Tg N yr^{-1} and 32 Tg N yr^{-1} with and without correction for non-natural land, respectively. Considerable uncertainty exists regarding true rates of BNF in natural ecosystems. The LPJ-GUESS prediction is substantially lower than estimates by Cleveland et al. (1999), The latter rate is considerably lower than observation-based estimates of potential N fixation, which lie in the range of 100–290 Tg N yr^{-1}. However, recent studies suggest that this may have been an overestimation (Sullivan et al., 2014; Vitousek et al., 2013). Nevertheless, also compared

20   to these new estimates, our global BNF rate is relatively low. This may be, and is due to a known discrepancy between the evapotranspiration (AET) simulated by LPJ-GUESS (which falls within the range of observational estimates) and the values on which the N fixation AET relationship is based (Smith et al., 2014). The low simulated values of BNF are particularly relevant in for the tropics, where BNF provides the dominant source of N input for ecosystems.

4.1.2   Climate

25   Predicted N leaching response to climate is complicated by several issues. First, temperature and precipitation change simultaneously, since for consistency we chose to treat climate as a single factor in the experiment. Furthermore, while temperature change is globally relatively uniform and mostly positive (supplemental Figure S2), precipitation is spatially highly variable, showing both increases and decreases over the 20th century (supplemental Figure S3). In many regions, the distribution of precipitation over rain days has changed as well (not shown), influencing the intensity of precipitation and

30   thereby percolation fluxes.

Second, both variables affect various ecosystem processes that influence N leaching in opposing directions. Temperature influences both N mineralization and vegetation productivity, while precipitation stimulates both soil water fluxes and N input by BNF, which is linked to evapotranspiration in the model. Both variables influence fire probability, which is regionally very relevant.

5   Globally, climate change has a positive effect on N leaching (Figure 12~~Figure 11~~), mainly due to an increase in net N mineralization caused by warming (supplemental Figure S22~~19~~), and a small and mostly negative effect on productivity (supplemental Figure S15~~9~~). Particularly in regions with high soil organic carbon storage and regions that are N rich (Western Europe; Russia; Canada) climate change has a strong positive effect on predicted N leaching. Negative N leaching response occurs in regions with stimulated productivity, or reduced precipitation in combination with high N deposition

10   (Northern India). Climate change also stimulates fire occurrence.

Similar to the LPJ-GUESS results, findings of observational studies on the effect of temperature on N leaching vary, depending on site conditions. Soil warming experiments generally show a stimulation of vegetation productivity, which is usually attributed to increased N availability caused by stimulated mineralization (Melillo et al., 2011; Rustad et al., 2001). Studies that report leaching fluxes support the LPJ-GUESS results in that N rich sites usually show a clear increase in N

15   leaching (Joslin and Wolfe, 1993; Lükewille and Wright, 1997; Schmidt et al., 2004), while N poor sites have a less strong response (Schmidt et al., 2004). It should be noted, however, that soil warming experiments may not be fully compatible with our results since they only account for the effect of increased N availability on vegetation productivity but not direct effects of increased temperature on plants.

Global long-term trends in N leaching do not appear to be related to precipitation in our study. At the regional scale,

20   however, changes in precipitation can be quite important for N leaching, as shown by the factorial experiment (section 3.2.2). Furthermore, spatial patterns of N leaching are strongly linked to soil water fluxes, which relate directly to precipitation, as illustrated by the strong relationship between runoff and N leaching on log-log scale (supplemental Figure S21~~18~~). This relationship compares well with findings of Lewis et al. (1999)~~Lewis et al., (1999)~~.

4.1.3   Atmospheric CO_2 concentration

25   LPJ-GUESS predicts a considerable negative response of N leaching to changes in atmospheric CO_2 concentration (Figure 11~~Figure 10~~c and Figure 12~~Figure 11~~). Strong reductions occur in regions which have high N availability, either due to N deposition (temperate biomes) or N fixation (tropics). Furthermore, CO_2 also stimulates litter production, which tends to increase fire occurrence, resulting in volatilisation N losses, which lead to a compensatory reduction in leaching in semi-arid biomes. Particularly in the tropics LPJ-GUESS predicts a strong GPP response to CO_2 increase (supplemental Figures~~.~~ ~~S15,~~

30   S19~~6~~). Due to the lack of CO_2 enrichment experiments in the tropics, little is known about the CO_2 response of tropical forests (Hickler et al., 2008); hence it is unclear to what is extent this behaviour is realistic. It is likely that in reality

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phosphorus availability limits GPP response (Wang et al., 2010), which would presumably reduce the negative effects on N leaching.

Although there is a large body of literature on the effects of CO_2 enrichment on ecosystems, few studies report N leaching. Several studies found reductions in N leaching under elevated CO_2 (Hagedorn et al., 2000; Hungate et al., 1999; Johnson et al., 2004) but absence of response has been reported as well (Larsen et al., 2011). In addition to enhancing plant productivity, elevated CO_2 tends to increase water use efficiency (Dekker et al., 2016), resulting in a reduction in transpiration. This stimulates runoff and water leaching (Betts et al., 2007), which could be expected to affect N leaching.

4.1.4   Fire

LPJ-GUESS predicts that fire plays an important considerable role in the N budget of natural ecosystems (Figure 2Figure 2, Figure 3Figure 3; supplemental Figure S109). Globally, fires account for 33 % of the predicted total N loss in the period 1997–2006, approximately the same as the contribution of N leaching. Ecosystems with frequent fires are more N limited and leach less N in the model predictionssimulations. This agrees with field studies that have found that fires lead to decreased N leaching on longer time scales (>3 years) (Johnson et al., 2007). Prescribed fires have been proposed as a measure to improve surface water quality (Fenn et al., 1998; Johnson et al., 2008). On shorter time scales, however, fire may enhance leaching due to mineralization of fire-induced litter fall (Alexis et al., 2007; Johnson et al., 2008). Furthermore, at burned sites original vegetation is often replaced by N fixing plants, which quickly replenish lost N (Johnson et al., 2008). Neither of these processes is represented in LPJ-GUESS.

If we adjust the LPJ-GUESS prediction of global N fire loss for the fraction of non-natural land we find an estimate of 16.7 Tg N yr^{-1}. This compares well with observation based estimates. For example, according to the Global Fire Emissions Database (GFED4; van der Werf et al., 2010), 16.4 Tg N yr^{-1} was emitted from natural fires globally ($N_2O + NH_3 + NO_x$) in 1997–2006. Schultz et al. (2008) reported a somewhat higher value for the 1990s: 24 Tg N yr^{-1} ($NH_3 + NO_x$). Although uncertainty regarding N losses by fire is large (Gruber and Galloway, 2008), the general agreement with observations strengthens our finding of the important role of fire in the terrestrial N budget. Specifically for savannah ecosystems, the influence of fire on N dynamics is further supported by field and remote sensing studies (Veldhuis et al., 2016; Chen et al., 2010).

It should be noted that part of the emitted N from fire is quickly returned to the land surface by atmospheric deposition. This is not accounted for in our study, since we prescribed N deposition, based on the dataset of Lamarque et al. (2013), which is derived from simulations with an ensemble of atmospheric chemistry models. The N emission sources used to drive these models include natural fires (Lamarque et al., 2010). However, while global total N emission by fire is comparable to the estimate of LPJ-GUESS (13.3 Tg N yr^{-1}), the spatial patterns of these emissions (supplemental Figure S11) differ considerably from those predicted by LPJ-GUESS (supplemental Figure S10). but it is unknown whether these agree with

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the fires predicted by LPJ-GUESS in terms of pattern and magnitude.A more detailed modelling study that accounts for local recycling of N emitted by fire is needed in order to draw stronger conclusions regarding the role of fires.

4.2   Ecosystem N status

We have used the N limitation factor, which reflects the reduction in photosynthetic capacity due to N limitation, as a proxy for N status (Figure 5Figure 5). While this quantity is in general correlated to N input, its spatial variability appears to be more strongly related to ecosystem type (supplemental Figure S8). The savannah, grassland, boreal forest, and tundra biomes show strong limitation, while for example tropical rainforests are generally closer to saturation. This agrees well with observational findings on the prevalence of N limitation in natural ecosystems (Vitousek and Howarth, 1991). Apart from differences in climate zone, the relationship between biome and N status is the result of the interaction of several PFT specific interactions. Grass PFTs have higher productivity, resulting in a higher N demand and will thus more quickly experience N limitation. In LPJ-GUESS tundra vegetation is represented by C3 grass, which explains the strong N limitation for this biome. Ecosystems dominated by Ggrass PFTs also experience more fire, which exacerbates N limitation in semi-arid regions. Phenology plays a role as well—regions with a short growing season due to seasonally low temperatures or drought are less efficient in retaining N.

For similar reasonsreasons, the relationship between N status and N leaching is also confounded. Although in general regions with high N status tend to lose more N by leaching according to our simulations (supplemental Figure S7), variability is large, due to the interplay of natural and ecological processes. For example, several arid regions (e.g. Chinese deserts) have generally high N status but leaching rates are quite low due to low water fluxes. On the other hand, in N limited ecosystems significant leaching can still occur (Figure 6Figure 6), demonstrating that there are limits to the efficiency of plants to retain N. To some extent these results contradict the conceptual model of N saturation proposed by Aber et al. (1998, 1989) which states that N leaching occurs at significant rates only when an ecosystem is fully N saturated. This view has previously been criticized by Lovett and Goodale (2011) who pointed to studies that found that N limited ecosystems can in fact leach considerable amounts of N.

Thus, N limitation of vegetation as a proxy for N status is of limited value for predicting N leaching in a global setting. Conversely, the ratio of N leaching :to N input may be a useful complementary indicator for N status. Ignoring high latitude regions, this quantity is relatively high in regions with high N inputs (e.g. Western Europe, North East USA, South East Asia, and the tropics; Figure 4Figure 4b), which agrees with observational findings that the proportion of N lost by leaching increases with N deposition (Aber et al., 2003; Fenn et al., 1998).

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4.3 Comparison of N cycling predictions with other ~~large scale~~large-scale studies

As discussed in section 3.1.3, there is good spatial agreement with N leaching rates of Beusen et al. despite the differences between the models. This ~~agreement~~ resemblance likely results at least partially from similarities in the N deposition maps used as input (notwithstanding Indonesia, see section 3.1.1). Although different data sources were used (Lamarque et al., 2013 for our study; Dentener et al., 2006 for Beusen et al.), there is considerable agreement between the two. Conversely, earlier in the 20th century N deposition input match~~es~~ less well, as does the predicted N leaching (supplemental Figure S1~~2~~5b). ~~This supports our finding that N input, and specifically N deposition, is in general the dominant factor driving spatial and temporal differences in N leaching rate.~~ Nevertheless, given the high uncertainty associated with these estimates, the good agreement for contemporary conditions is encouraging.

As discussed in section 2.1.2, the representation of denitrification in the version of LPJ-GUESS used in this study is based on a simple empirical relationship (1 % of daily N mineralization). While N loss due to denitrification is highly relevant for N leaching, accurate estimation of its contribution to the total N budget at large spatial scale remains a considerable challenge, both with observation-based approaches and mechanistic models. This is demonstrated by the disagreement between different estimates of f_{denit}, depicted in Figure 10. The zonal means show somewhat better agreement, but in general the difference between the LPJ-GUESS predictions and two other datasets is smaller than the difference among the observation-based estimates themselves. Nevertheless, certain observed patterns are reproduced by LPJ-GUESS, such as the relatively large contribution of denitrification in arid regions, caused by low percolation rates, and a tendency towards lower contribution in regions with high N deposition. An exception are the wet tropics, for which LPJ-GUESS predicts a lower contribution of denitrification compared to both observation-based datasets. A possible explanation for this mismatch is the fact that in wet regions high moisture availability creates conditions favourable for denitrification. In LPJ-GUESS, N leaching rates are more strongly linked to moisture availability (via drainage fluxes) than denitrification rates, resulting in a comparatively large predicted contribution of leaching.

After correction for non-natural land cover, based on the land-use dataset of Hurtt et al. (2011)~~Hurtt et al., (2011~~ (see section 2.3)~~)~~, we estimate a global N leaching rate of 18.5 Tg N yr^{-1} from natural ecosystems for 1997–2006. Although a number of studies have quantified terrestrial N export, most of these do not distinguish contributions from different land cover types. The studies that did report the contribution of natural ecosystems found rates of a magnitude comparable to our estimate. For example, Zaehle et al. (2010) found a substantially higher value of 27 Tg N yr^{-1} for the 1990s based on a simulation with the OCN land surface model. The difference with our study is at least partially explained by their higher estimate of biological N fixation (107.8 Tg N yr^{-1}). Recently, Nevison et al. (2016) reported N export (leaching + surface runoff) for 1995–2005, from a simulation with the Community Land Model with natural vegetation only. As a rough correction for non-natural land,

we may multiply their estimate (10.6 Tg N yr^{-1}) by 0.64, the ratio of our rate and the predicted N leaching for a completely natural world (28.6 Tg N yr^{-1}). This yields a value of 6.8 Tg N yr^{-1}, which is considerably lower than other published rates, including ours. The authors acknowledged that this is likely an underestimation caused by too high denitrification rates.

4.4   Model limitations

The LPJ-GUESS model is part of the state of the art of global ecosystem modelling and compares favourably with other models in model-data comparison studies (Zaehle et al., 2014). Furthermore, although the latitudinal pattern of GPP shows some mismatch with data-driven estimates (section 3.1.3), overall, agreement is sufficient for the purpose of this study. Nevertheless, uncertainties in modelling N cycling at the global scale are in general high. Competing model representations yield divergent results (Zaehle and Dalmonech, 2011), and available observations are currently not sufficient to identify best parametrizations. We will discuss several specific model limitations of LPJ-GUESS (also present in other models) that are particularly relevant for N leaching.

An important source of uncertainty stems from the representation of biological N fixation. As mentioned previously, BNF predicted by LPJ-GUESS is 3–9 fold lower than observation based estimates (Cleveland et al., 1999), which may be expected to cause underestimation of leaching. Furthermore, in LPJ-GUESS BNF occurs passively without explicit link to vegetation productivity or ecosystem N status. In reality plants are believed to exert some control on BNF rates based on the balance between N demand and availability (Vitousek et al., 2002). Since there is a considerable energy cost involved, plants tend to downregulate BNF under N rich conditions in favour of other pathways such as mycorrhizal, or passive uptake (Houlton et al., 2015). Similarly, N resorption from leaves before senescence—assumed a fixed fraction of 50 % in LPJ-GUESS—tends to be lower under N rich conditions. More mechanistic descriptions of plant N acquisition have been proposed (Brzostek et al., 2014), which account for these feedbacks, and could potentially result in improved prediction of ecosystem N cycling and leaching under varying conditions.

In the current version of LPJ-GUESS all mineral nitrogen forms are lumped into a single pool. In reality, however, inorganic nitrogen in soils exists in a range of chemical forms, most importantly nitrate (NO_3^-) and ammonium (NH_4^+). The behaviour of these two species in soil differs considerably (Butterbach-Bahl et al., 2011). Ammonium is much less susceptible to leaching since soils generally have a much higher capacity for retaining cations than anions. As a result, inorganic N losses occur mainly in the form of nitrate and its formation by nitrification is an important control for leaching losses (Zhang et al., 2016). Since plants and microbes preferentially take up ammonium over nitrate, nitrification rates are usually low in N limited ecosystems. Hence, it is possible that leaching losses are overestimated in N limited ecosystems, which may explain the mismatch with observations (supplemental Figure S20) at low N deposition rates. Transformation of ammonium to nitrate by nitrification, and vice versa by denitrification is associated with gaseous losses. The rates of these processes are highly dependent on substrate concentrations and soil aeration status. Particularly denitrification can be a

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considerable loss term in the N budget under anaerobic conditions (Bouwman et al., 2013a). The relatively simple representation of denitrification ~~Since LPJ-GUESS models gaseous losses as a fixed fraction~~ (1 %~~)~~ of gross N mineralization~~)~~, in LPJ-GUESS roughly reproduces global spatial patterns of relative N loss by this process (sections 3.1.3 and 4.3), but it is possible that predictions of N leaching are ~~it is likely to~~ overestimate~~d~~ leaching losses in soils with high N

5   availability under wet conditions. Currently, LPJ-GUESS is being updated with an improved description of soil N cycling based on the approach of Xu-Ri and Prentice (2008) that accounts for different mineral N species and transformations between them.

Finally, LPJ-GUESS predicts that dissolved organic N (DON) leaching is generally negligible compared to the overall N budget (supplemental Figure S6; globally 0.18 % of total N loss for 1997–2006) which likely represents an underestimation.

10   Observations based on river concentrations indicate that DON loss can be a significant component of ecosystem N export, particularly in N limited regions (Perakis and Hedin, 2002). In process-based modelling studies DON leaching has been largely ignored as a significant N sink, which is increasingly recognized as a limitation (Nevison et al., 2016).

5   Conclusions

The factorial experiment with LPJ-GUESS allows us to disentangle the effects of changes in N deposition, climate, and

15   atmospheric CO_2 concentration on N leaching. From a global perspective N deposition is the most important control of N leaching in our model simulations. Rising N deposition during 20th century has caused large increases in N leaching in many regions in the world. Rising atmospheric CO_2 and climate change, of secondary but not negligible importance, have a negative and a positive effect on N leaching, respectively. Although temporal trends are clear at the global scale, there are large regional differences, even when individual drivers are considered. This variability results largely from heterogeneity of

20   climate and N deposition changes and biome type, and causes complex spatial patterns when all three drivers are combined. These patterns would have been difficult to understand based on the true historical simulation alone. Ecosystem N status is more difficult to assess based on our results. Spatial patterns of N limitation on vegetation productivity are more strongly related to vegetation type than N input. Nevertheless, at the global scale N limitation is clearly decreasing and regions with highest N deposition are approaching N saturation.

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(a) Atmospheric N deposition 2000–2010 (kg ha^{-1} yr^{-1})

(b) Change relative to 1850–1860 (kg ha^{-1} yr^{-1})

Figure 1. Atmospheric N deposition for 2000–2010, (the last decadal interval of the ACCMIP historic N deposition data set (Lamarque et al., 2013). (a) Absolute rates. (b) Change relative to 1850–1860. For both figures the colour axis is cut off at approximately the 99 % quantile to improve readability.
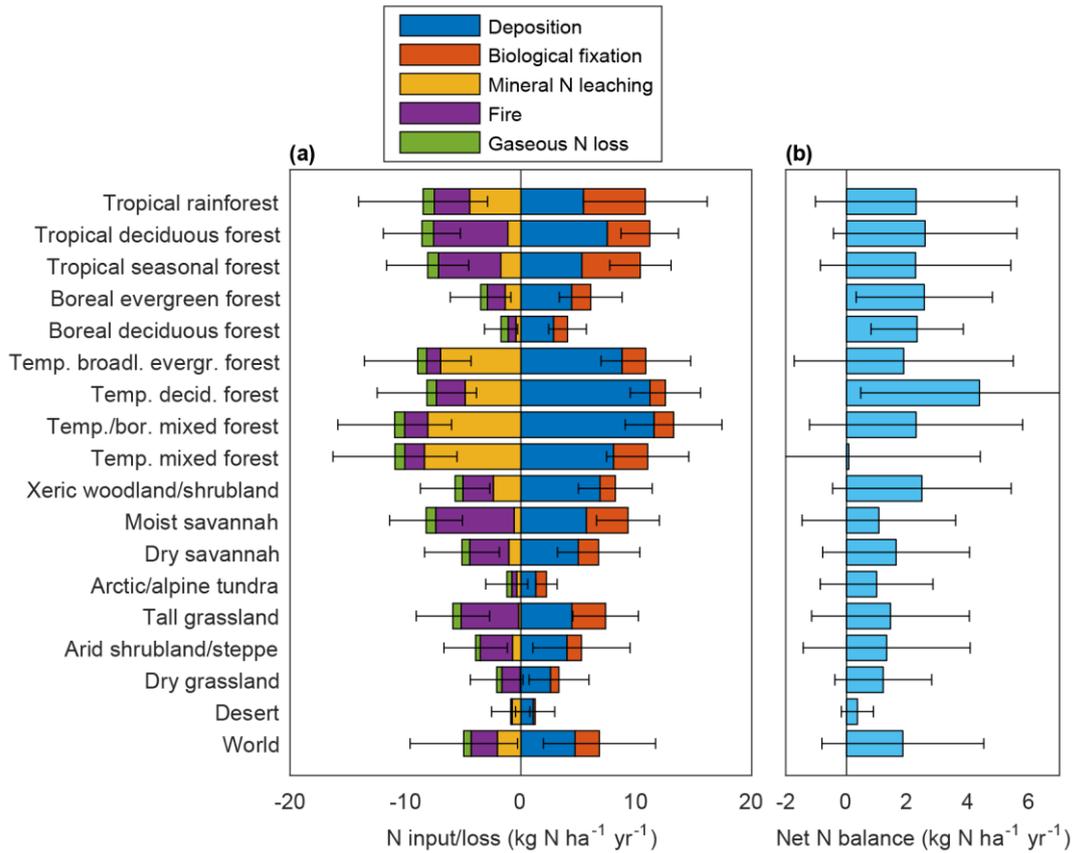
Figure 2. Ecosystem N budget per biome for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006. (a) Mean N input and loss fluxes; (b) Mean net ecosystem exchange of N. Organic N leaching (not shown) is negligible for all biomes. Error bars indicate one standard deviation among area-weighted grid cell averages for the target time period.
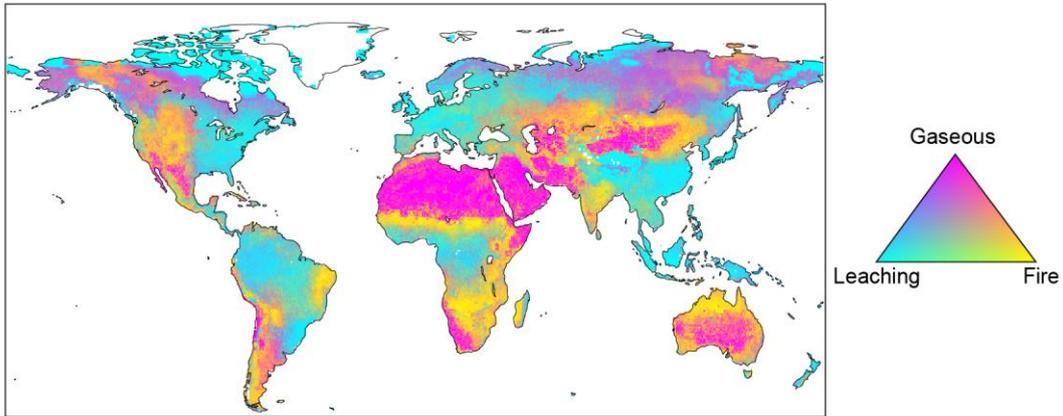
Figure 3. Relative contribution of leaching, gaseous loss, and fire to overall ecosystem N loss for the true historical simulation (+Ndep +clim +CO2) averaged over the period 1997–2006. The colours at the three corners of the triangle indicate 100 % N loss by the corresponding process.
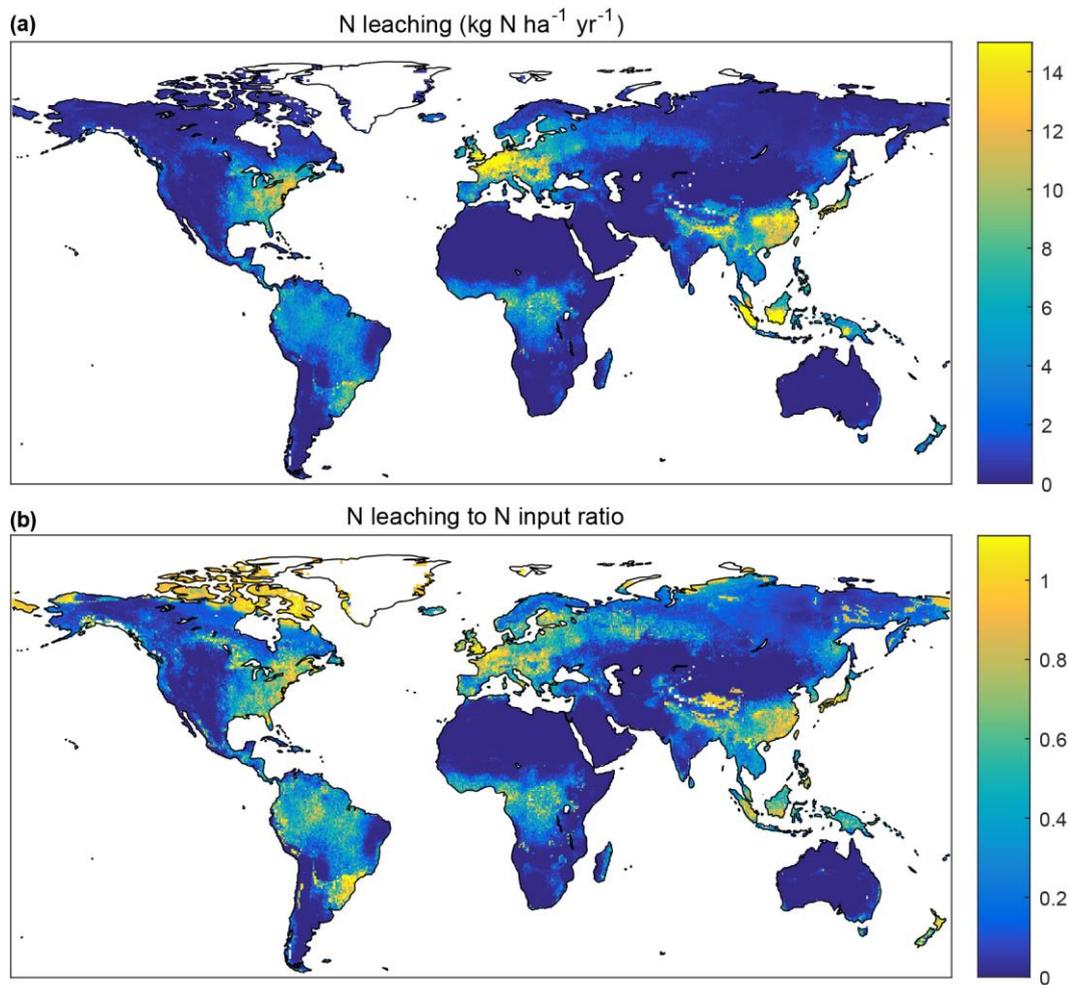
Figure 4. Mineral N leaching (kg N ha⁻¹ yr⁻¹) for the true historical simulation (+Ndep +clim +CO2) averaged over the period 1997–2006. (a) N leaching; (b) ratio of N leaching and N input (N deposition + BNF). For ~~readability~~readability, **the colour axes have been cut off at the 99 % quantile.**
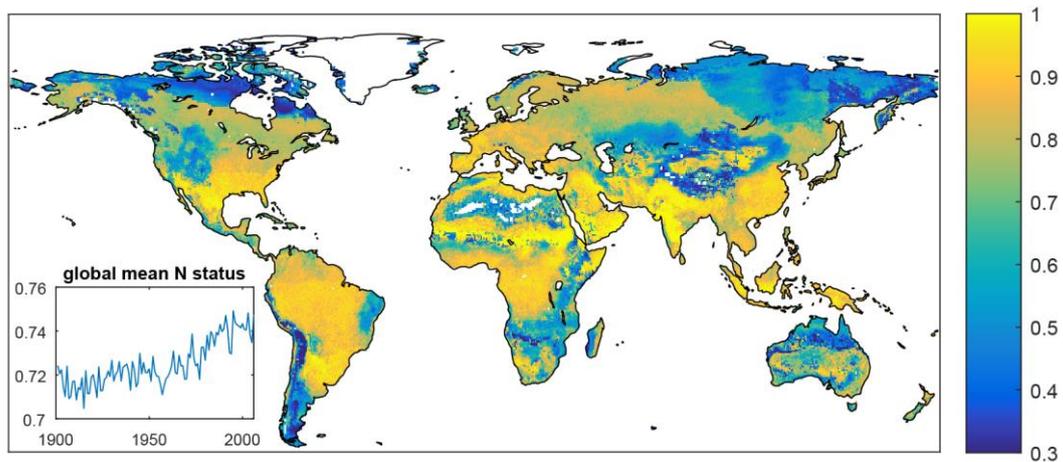
Figure 5. Ecosystem N status for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006. Lower values indicate stronger N limitation (see main text). The inset shows global mean N status over time during the simulation period.
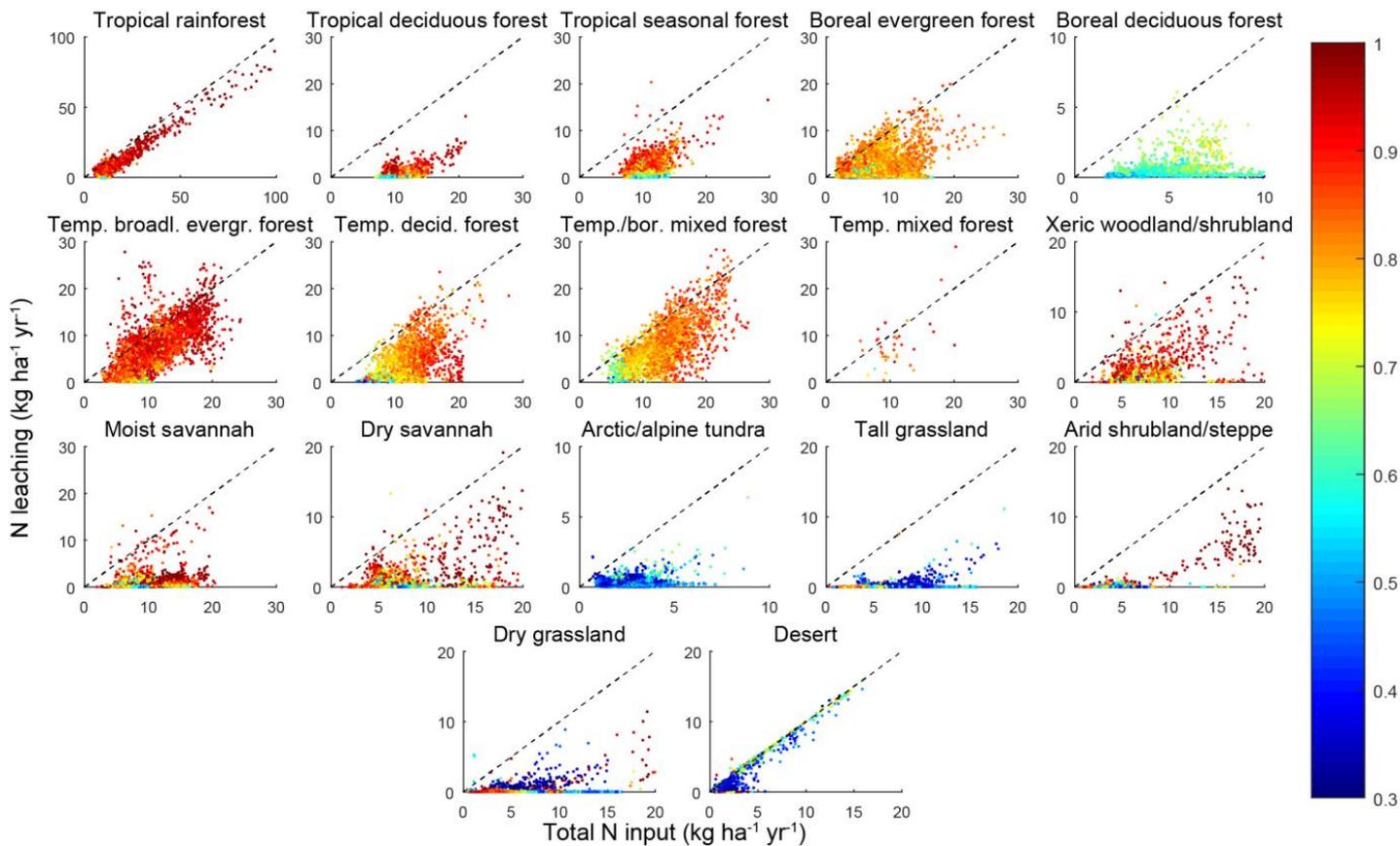
Figure 6. Mineral N leaching versus total N input (deposition + BNF) for the true historical simulation (+Ndep +clim +CO2) averaged over the period 1997–2006. Each point represents one grid cell. ~~Colors~~Colours indicate the mean N status. Dashed lines indicate the 1:1 relationship.
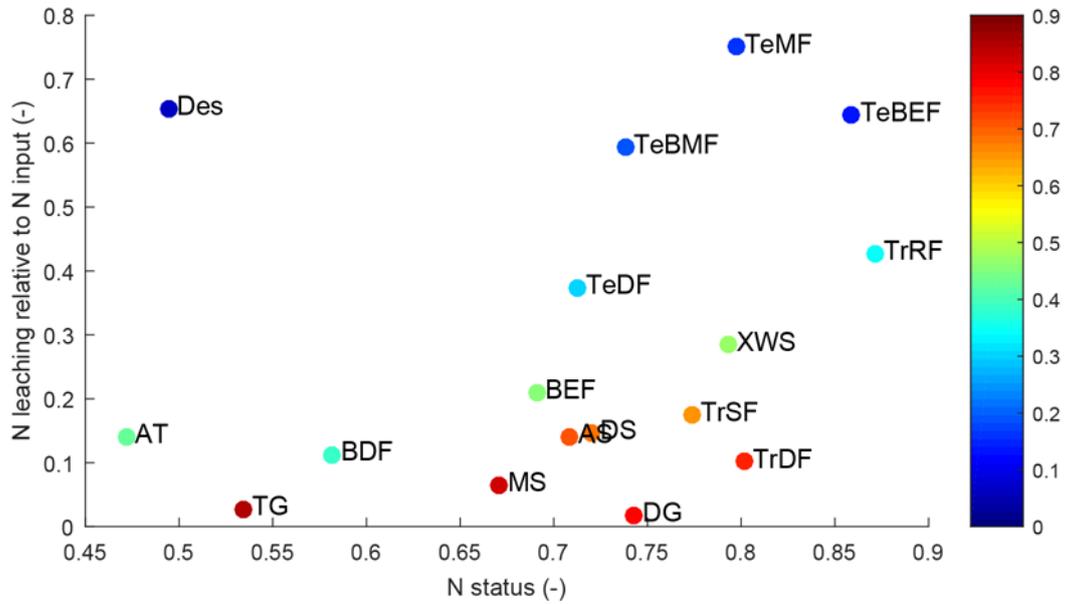
Figure 7. N leaching relative to total N input versus N status for the 17 biomes for the true historical simulation (+Ndep +clim +CO2). Colours indicate the mean fraction of N lost by fire. TrRF: Tropical rainforest, TrDF: Tropical deciduous forest, TrSF: Tropical seasonal forest, BEF: Boreal evergreen forest/woodland, BDF: Boreal deciduous forest/woodland, TeBEF: Temperate broadleaved evergreen forest, TeDF: Temperate deciduous forest, TeBMF: Temperate/boreal mixed forest, TeMF: Temperate mixed forest, XWS: Xeric woodland/shrubland, MS: Moist savannah, DS: Dry savannah, AT: Arctic/alpine tundra, TG: Tall grassland, AS: Arid shrubland/steppe, DG: Dry grassland, Des: Desert.
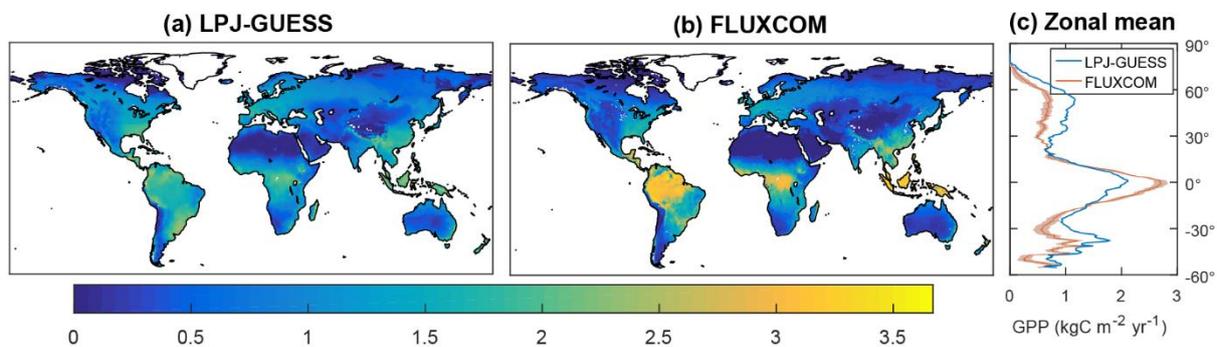
Figure 8. Mean gross primary productivity (GPP) for 1997–2006. (a) LPJ-GUESS GPP for the true historical simulation (+Ndep +clim +CO2). (b) FLUXCOM GPP (Jung et al., 2017), mean over six approaches. (c) latitudinal averages. The shaded area for FLUXCOM indicates the 95 % confidence range over the six approaches.

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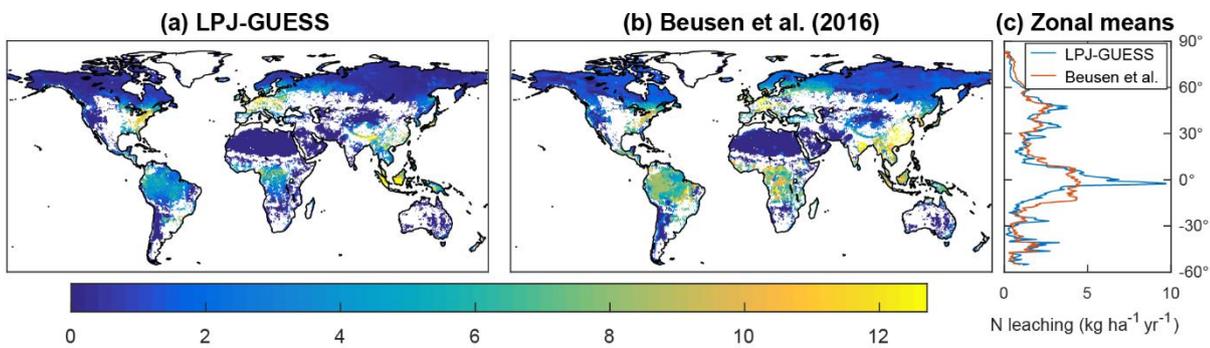
Figure 9. Mean mineral N leaching rate for 1999–2000 compared to estimates of Beusen et al. (2016). (a) prediction by LPJ-GUESS for the true historical simulation (+Ndep +clim +CO2). (b) estimate of Beusen et al. for natural ecosystems only. (c) latitudinal averages. The many missing values in the maps are caused by mask for natural land cover of Beusen et al. which was applied to both datasets (see section 2.4).
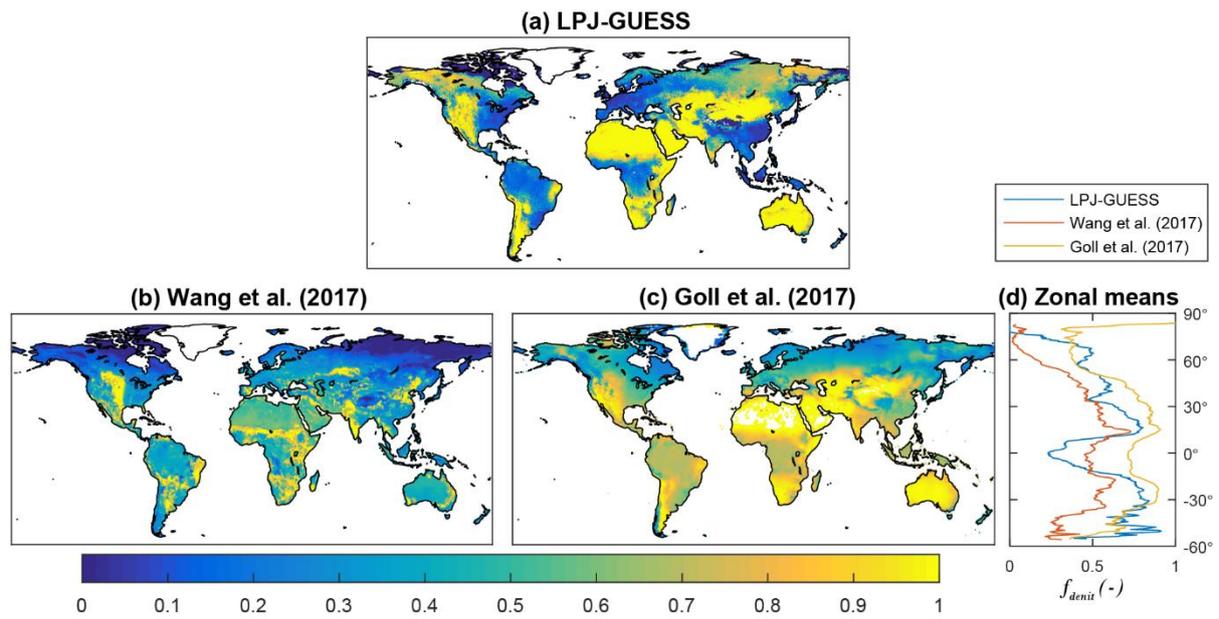
Figure 10. Relative contribution of denitrification to total soil N loss (excluding fire). (a) Prediction by LPJ-GUESS for the true historical simulation (+Ndep +clim +CO2), 1997–2006. (b) Estimate by Wang et al. (2017); (c) Estimate by Goll et al. (2017). (d) latitudinal averages.
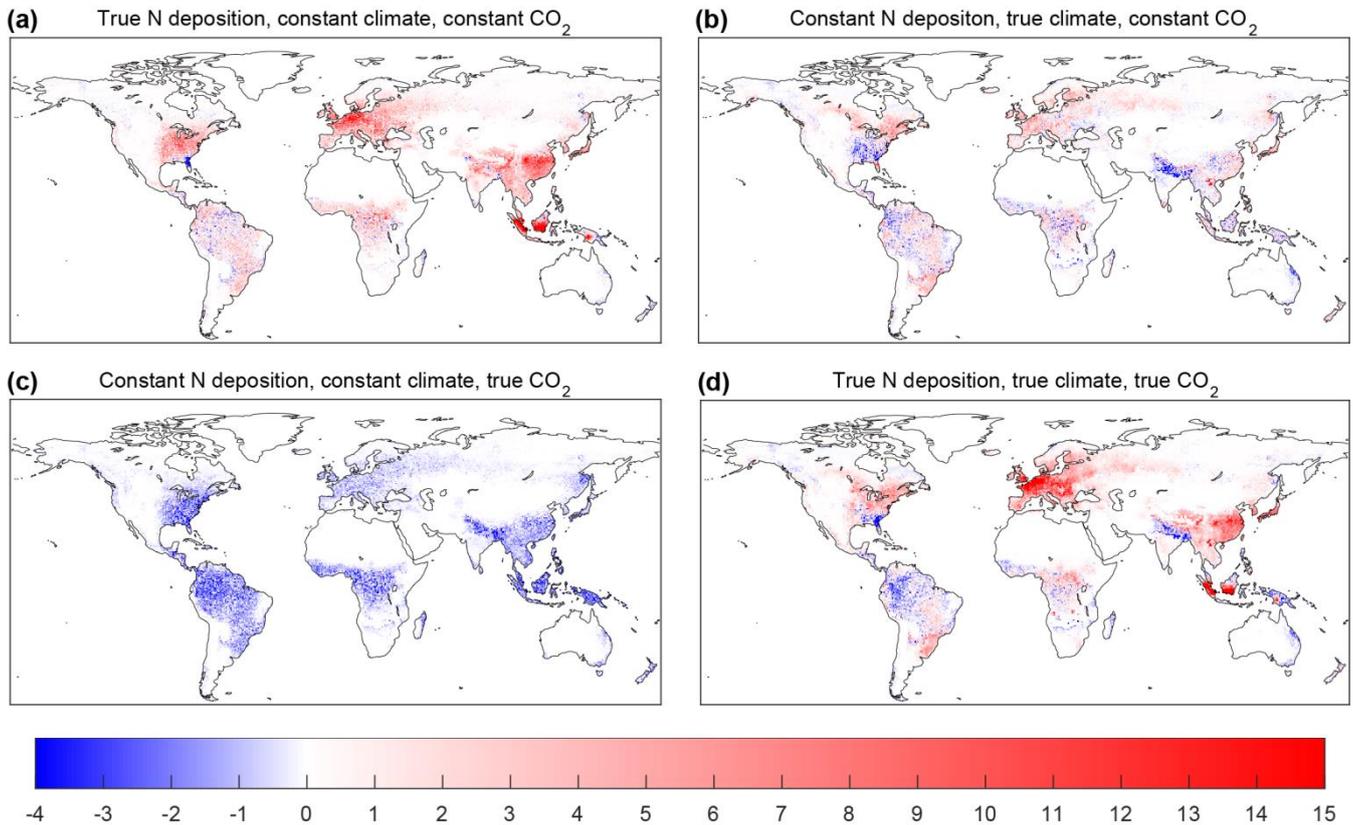
Figure 1110. Mineral N leaching difference relative to the control simulation (-Ndep -clim -CO₂) for the single factor runs and the true historical simulation. For ~~readability~~readability, the ~~color~~colour axis has been cut off at the 1 % and 99 % quantile over all graphs.
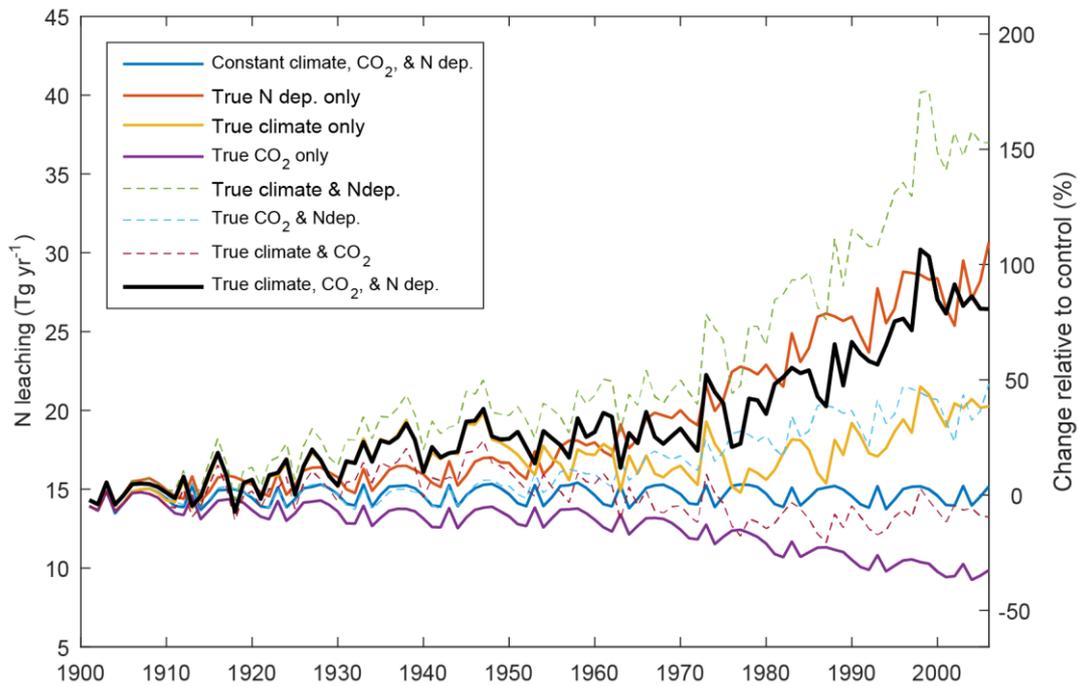
Figure 1211. Global total N leaching for the eight simulations, for a world with potential natural vegetation only. The ~~right hand~~right-hand side axis depicts change relative to the mean rate for the control simulation (-Ndep -clim -CO2).

Table 1. Simulation runs in the factorial simulation experiment. Note that climate comprises four variables (section ~~2.2.1~~2.3).

Label	N deposition	Climate	Atmospheric CO_2
-Ndep -clim -CO_2	constant	constant	constant
+Ndep -clim -CO_2	true	constant	constant
-Ndep +clim -CO_2	constant	true	constant
-Ndep -clim +CO_2	constant	constant	true
+Ndep +clim -CO_2	true	true	constant
+Ndep -clim +CO_2	true	constant	true
-Ndep +clim +CO_2	constant	true	true
+Ndep +clim +CO_2	true	true	true

Earth System Dynamics

Supporting Information for

Nitrogen leaching from natural ecosystems under global change: a modelling study

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Contents of this file

Text S1. Description of LPJ-GUESS

General description

LPJ-GUESS (Lund-Potsdam-Jena General Ecosystem Simulator; Smith et al., 2001) simulates vegetation dynamics and biogeochemical fluxes of C and N in terrestrial ecosystems and employs generalized biome- or global-scale parameterizations of component ecosystem processes, allowing it to be employed without recalibration globally or for any large region. It is forced by climate variables, CO_2 concentration and N deposition and runs with a daily time step, except for C allocation, vegetation dynamics, and disturbances, which are resolved annually. Our simulations focused on natural vegetation, i.e. croplands were not considered. Eleven plant functional types (PFTs) were included, representing vegetation in temperate, tropical, boreal, and grassland biomes. The model predicts the occurrence of each PFT based on bioclimatic limits and competition with other PFTs for light and soil resources. Contrary to most global ecosystem models, LPJ-GUESS explicitly represents the age distribution of woody PFTs. The model simulates trees of different cohorts (age classes) which are each represented by an average individual for each age class of each of a number of co-occurring PFTs. Mortality and establishment of the individuals are implemented in a stochastic fashion, as are fire and other disturbances (see below). Sub-grid variability resulting from landscape heterogeneity and differences in disturbance history are accounted for by simulating a predefined number of replicate "patches" (area 0.1 ha) per grid cell. The conditions for all patches within a grid cell are identical but differences arise from the stochastic calculations.

Within each patch LPJ-GUESS simulates fluxes of C, water and N, in vegetation and soil based on descriptions of various processes, including photosynthesis, plant C allocation, autotrophic respiration, evapotranspiration, percolation, lateral runoff, and soil carbon cycling. Soil carbon cycling is simulated using a scheme based on the CENTURY model, as described by Smith et al. (2014) . Soil hydrology is represented using two soil layers of 0.5 and 1 m. Downward percolation is simulated with a leaky bucket scheme (Gerten et al., 2004). Available water capacity (AWC) and a hydraulic conductivity are derived from sand, silt and clay fraction, as described in Olin et al. (2015). Water in excess of the AWC in the first and second soil layer is exported as surface runoff and interflow, respectively.

Fire is modelled stochastically according to the scheme described in Thonicke et al. (2001) ~~which models~~. Fire can occur when a fuel (litter) load of 200 g m^{-2} or higher is present. When this is the case, the probability of a fire ~~occurrence as a~~ occurring on a given day is a non-linear function of ~~above ground litter storage and~~the moisture content of the upper soil layer~~. When~~, serving as proxy for the litter layer. The actual occurrence of a fire is determined using a random number generator. If a fire occurs, a PFT dependent fraction of the biomass (50–90 %) is lost from the ecosystem. Additional disturbances, killing all vegetation, are modelled with a fixed expected return time of 100 years.

The simulation is initialized with a ~~500 year~~500-year spin-up to accumulate vegetation and soil C pools in equilibrium with the initial forcing. During this ~~phase~~phase, the model is forced by a trend-free time series (here 10 years; see below) of annually-varying inputs.

N cycling module

In LPJ-GUESS ecosystem N is present in vegetation biomass and in the soil in mineral and organic form. In the model version employed for our study, mineral soil N is represented by a single pool; i.e. different N species such as ammonium and nitrate, and transformation between these are not distinguished. Atmospheric N deposition is added to the soil mineral N pool. Biological N fixation (BNF) is calculated as a linear function of evapotranspiration (Cleveland et al., 1999) and added to the mineral N pool, up to a maximum pool capacity of 2 g N m^{-2}. Root uptake transfers N from the soil mineral N pool to vegetation on a daily time step. Plant N demand is driven by optimal leaf N content required for photosynthesis, computed based on the carboxylation capacity of Rubisco (V_{max}) that maximizes canopy-level net photosynthesis (Haxeltine and Prentice, 1996), given current atmospheric CO_2 concentration, temperature, soil water, and leaf area index (LAI). In addition to leaves plants require N for sapwood, heartwood, and roots. Plants also maintain an N store pool with a maximum size defined by biomass, leaf C:N ratio, and PFT type. Following Meyerholt and Zaehle (2015), the C:N ratios of non-leaf pools are fixed, which represents a modification to the model version described in Smith et al. (2014), where C:N ratios of non-leaf tissue were scaled based on leaf C:N. Plants take up N from the mineral soil pool and the N store in order to maintain optimal leaf N. If insufficient N is available the plant experiences N stress and V_{max} is reduced. To this end the model calculates an "N limitation factor" equal to the ratio of the true V_{max} and the V_{max} in absence of N limitation (both without water limitation). Additionally, different PFT cohorts compete for N, where the competitive strength of an individual is determined by its root biomass, the combined C:N ratio of roots and leaves, and growth form, with grass PFTs being more competitive than tree PFTs.

N stored in vegetation is returned to the soil in organic form in conjunction with biomass turnover due to senescence, mortality, and disturbance. Retranslocation transfers 50 % of leaf and root N to the N store pool, up to a maximum capacity, prior to turnover. Litter and soil organic matter (SOM) dynamics follow the CENTURY model (Parton et al., 1993), which includes nine litter and SOM pools, and two microbial pools. The C:N ratio of the slowest SOM pool (passive) is fixed, while the other pools vary depending on litter N content and soil available N. During decomposition N is transferred to or from the mineral N in order to maintain the C:N ratio of the pools. If insufficient N is available decomposition rates are reduced. Additionally 1 % of the daily N mineralization is lost, representing gaseous N loss during nitrification and denitrification. Organic N leaching occurs as a fraction of the soil microbial N pool, determined by the percolation rate and the sand fraction. Mineral N leaching is calculated as a fraction of the mineral N pool equal to the relative water loss by percolation and interflow. Surface runoff does not cause N loss. Finally, fire events cause loss of vegetation N which is assumed to be emitted in gaseous form.
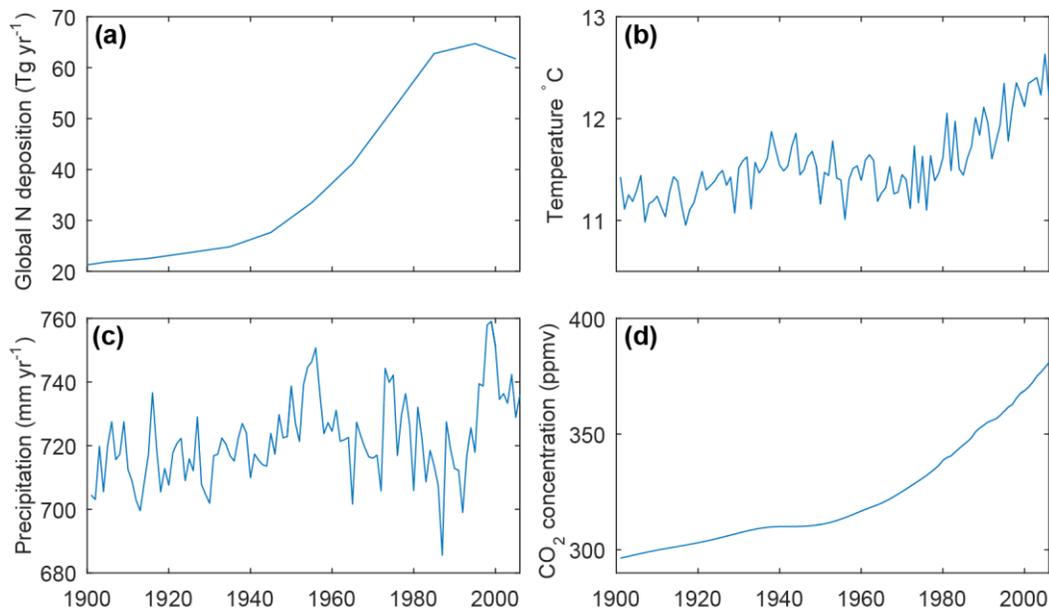
Figure S1. Change of model drivers during the simulation period. (a) global total atmospheric N deposition; (b) global mean temperature; (c) global mean precipitation rate; (d) global mean atmospheric CO_2 concentration.
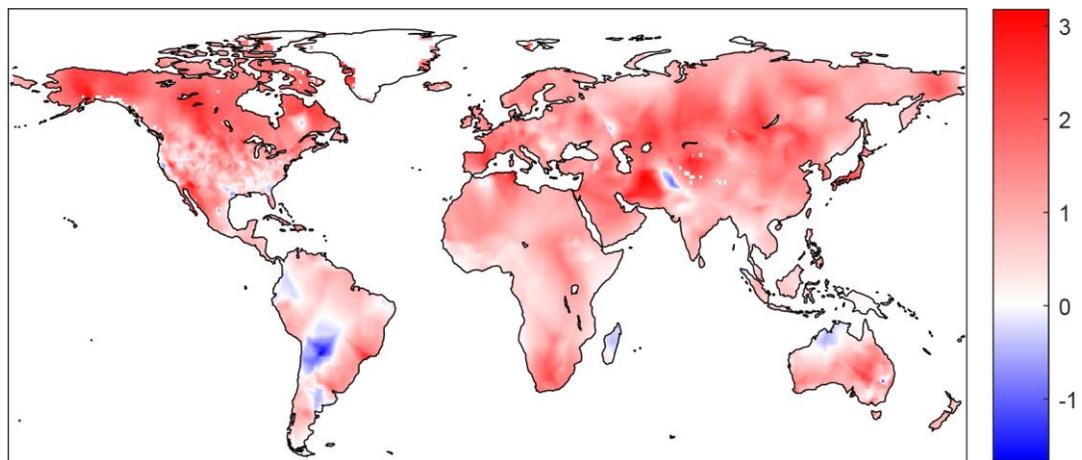

Figure S2. Mean temperature difference between 1997–2006 and 1901–1910 (°C).
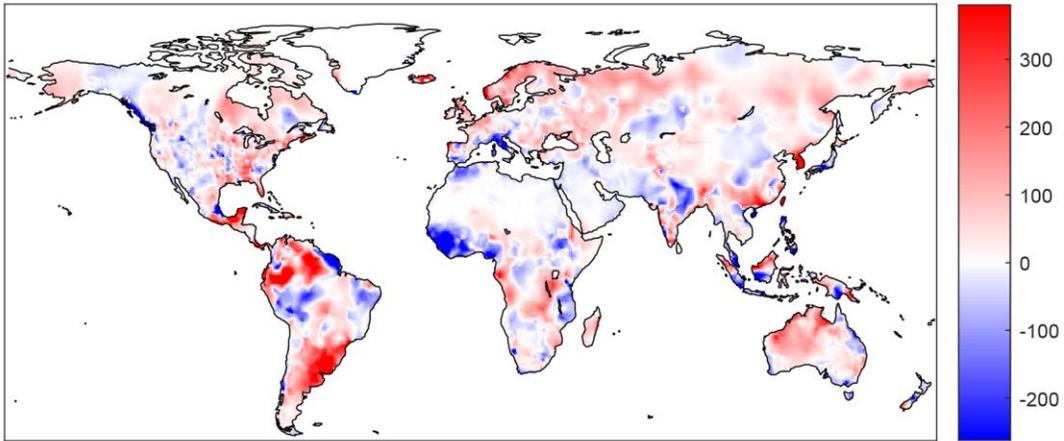
Figure S3. Mean precipitation difference between 1997–2006 and 1901–1910 (mm yr^{-1}).

Boreal decid. forest/woodland
Boreal evergr. forest/woodland
Temperate/boreal mixed forest
Temperate deciduous forest
Temperate broadl. evergr. forest
Temperate mixed forest
Tropical seasonal forest
Tropical rainforest
Tropical deciduous forest

Moist savannah
Dry savannah
Tall grassland
Dry grassland
Xeric woodland/shrubland
Arid shrubland/steppe
Desert
Arctic/alpine tundra

Figure S4. Biome distribution for the true historical simulation (+Ndep +clim +CO$_2$). Biome classes are derived from leaf area index of the PFTs averaged over the period 1997–2006 (Smith et al., 2014).
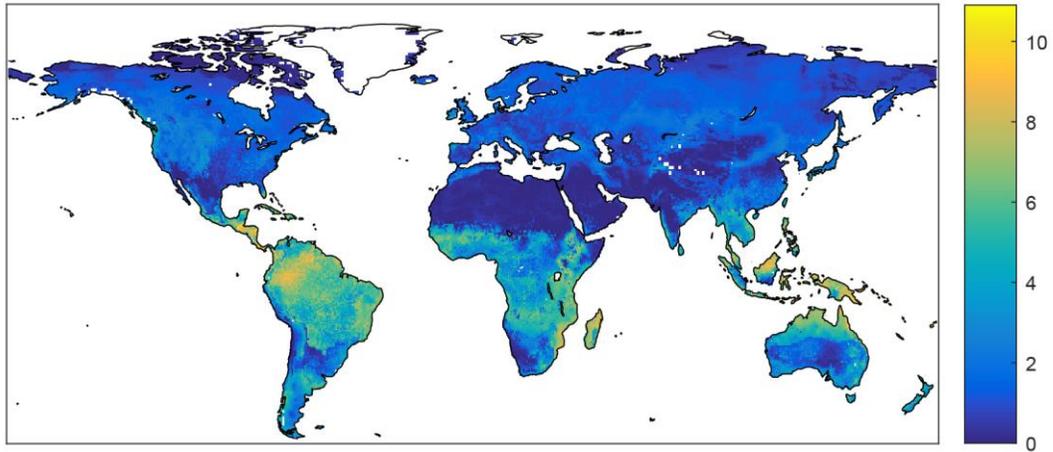
Figure S5. Biological N fixation (kg-N ha^{-1} yr^{-1}) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006.
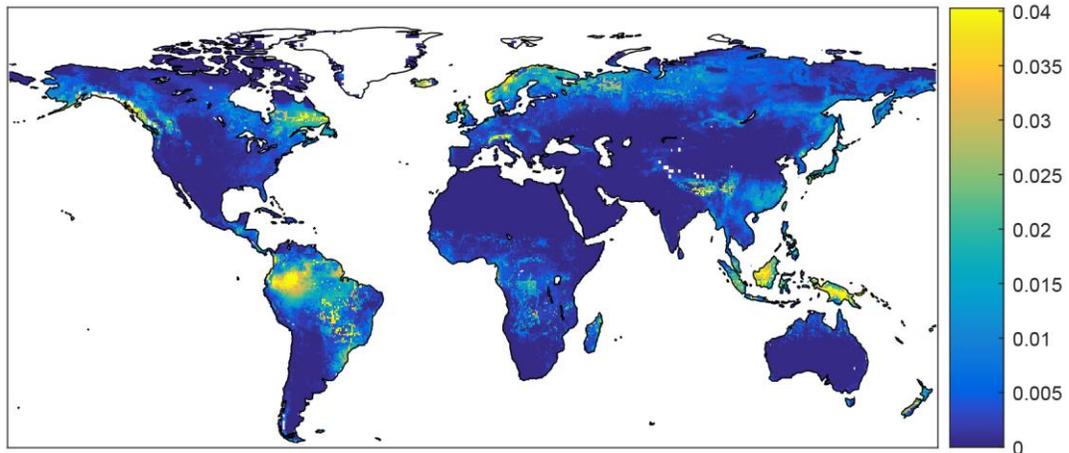
Figure S6. Organic N leaching (~~kg~~kg N ha^{-1} yr^{-1}) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006.
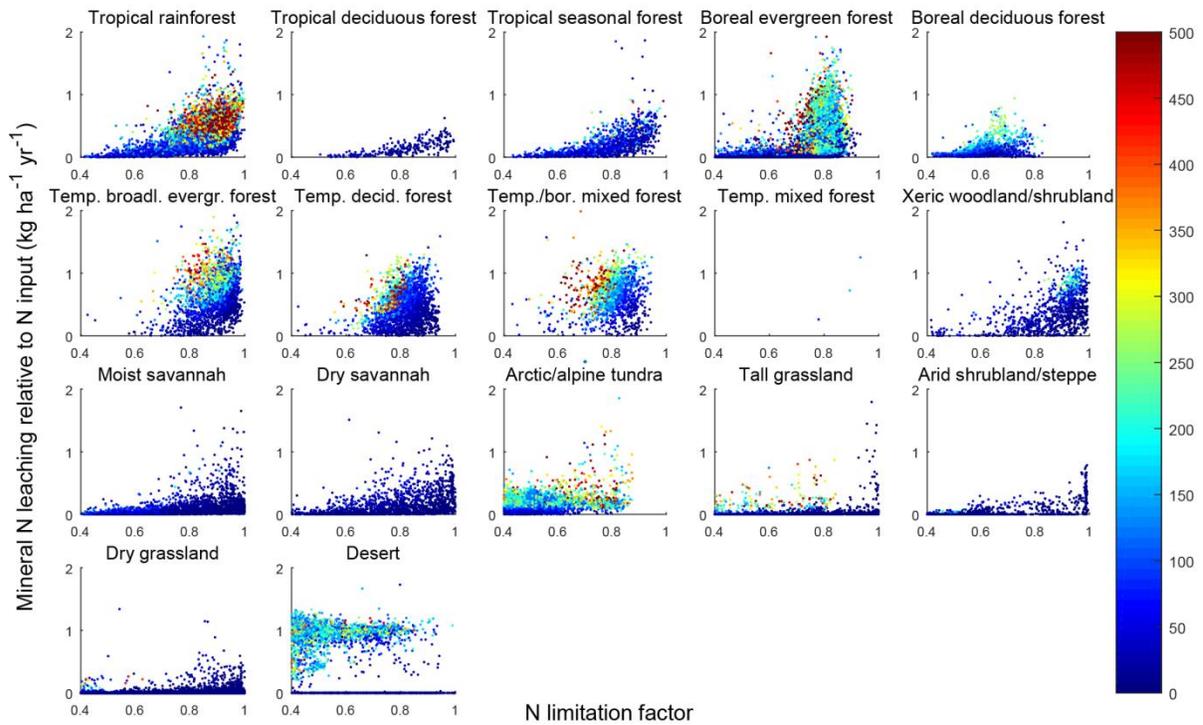
Figure S7. N leaching relative to N input vs N status for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006. Colors indicate the combined rate of interflow and percolation from the root zone (mm yr^{-1}; upper limit cut off to improve readability)
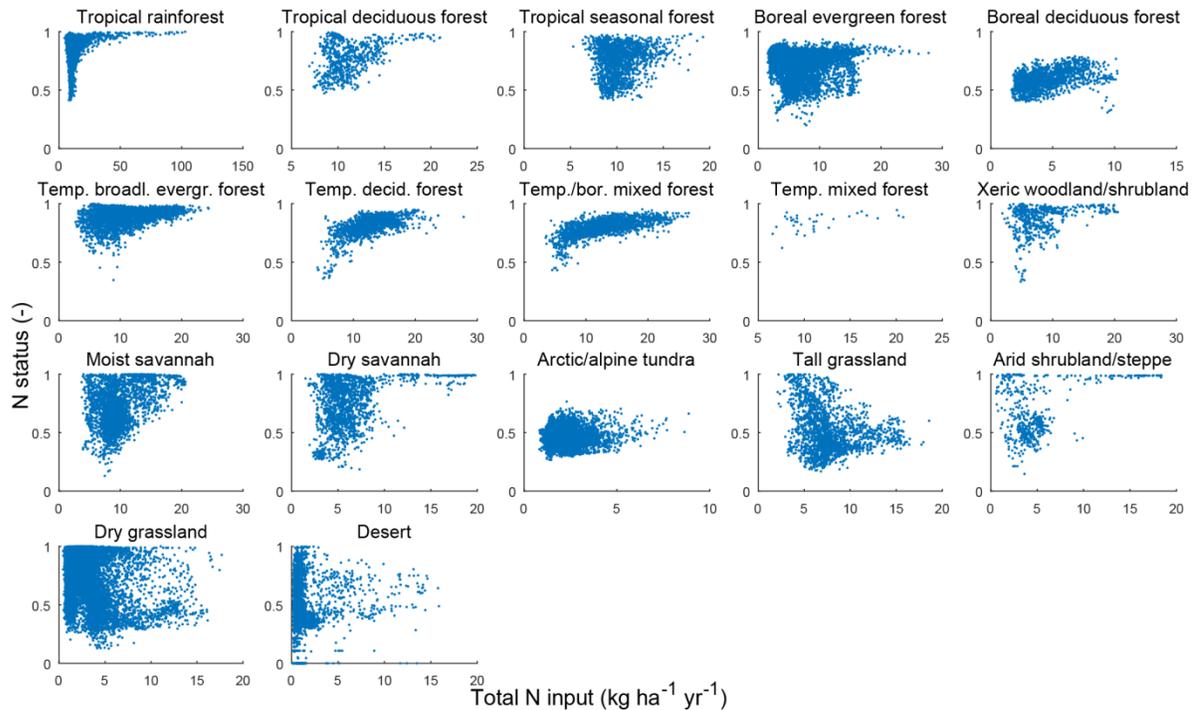
Figure S8. N status vs total N input (fixation + deposition) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006.

(a) Absolute (kg ha⁻¹ yr⁻¹)
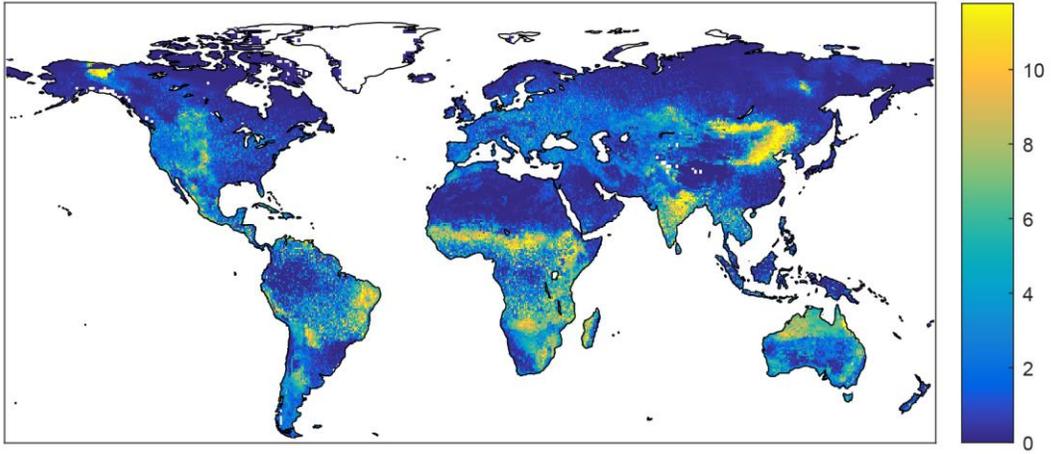
(b) Relative to N input
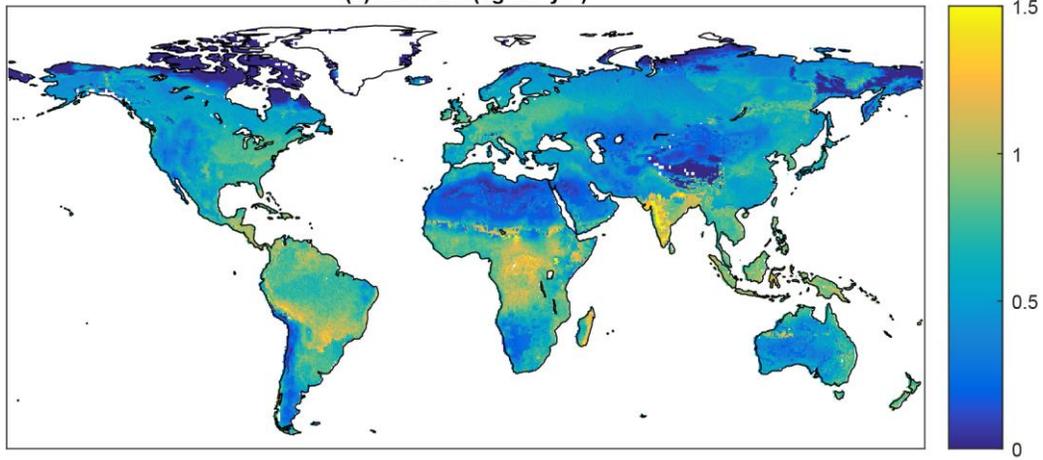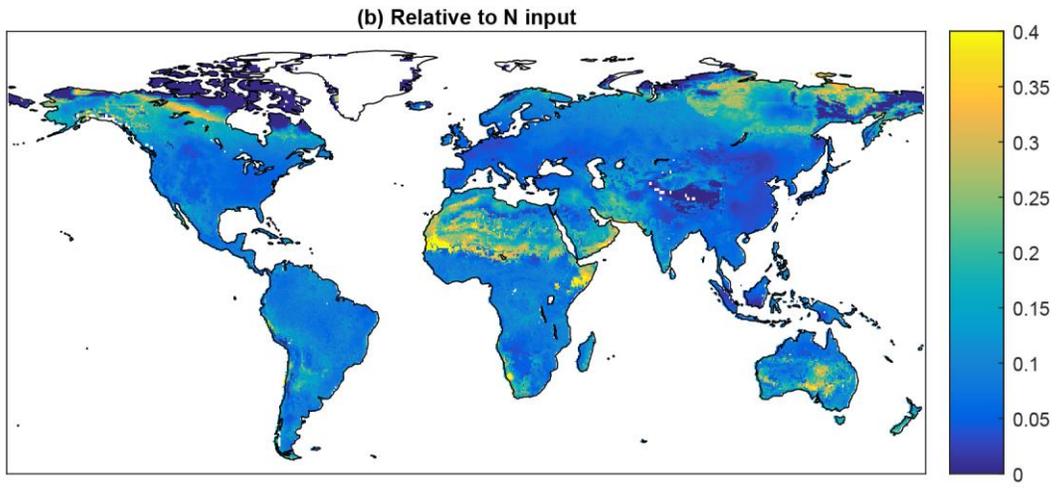
Figure S9. N loss due to ~~fire (kg ha⁻¹ yr⁻¹)~~ denitrification (kg ha⁻¹ yr⁻¹) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006. (a) Absolute; (b) Relative to total N input (deposition + fixation)

(a) Absolute (kg ha⁻¹ yr⁻¹)

(b) Relative to N input
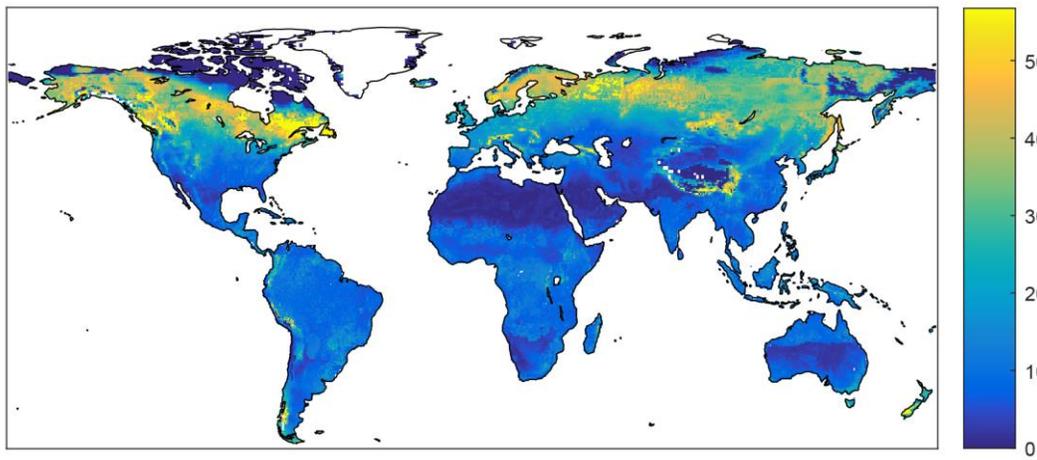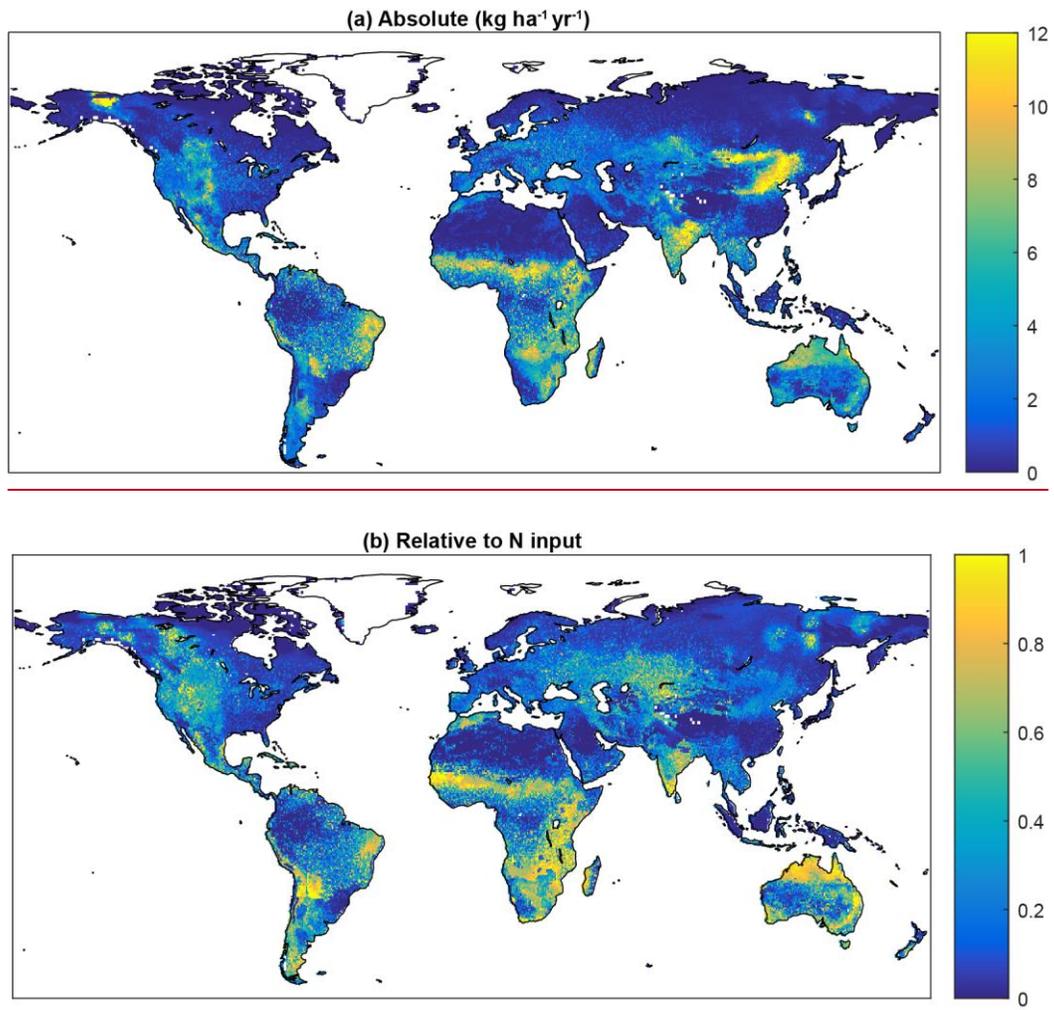
Figure S10. N loss due to fire (kg N ha^{-1} yr^{-1}) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006. (a) Absolute; (b) Relative to total N input (deposition + fixation)
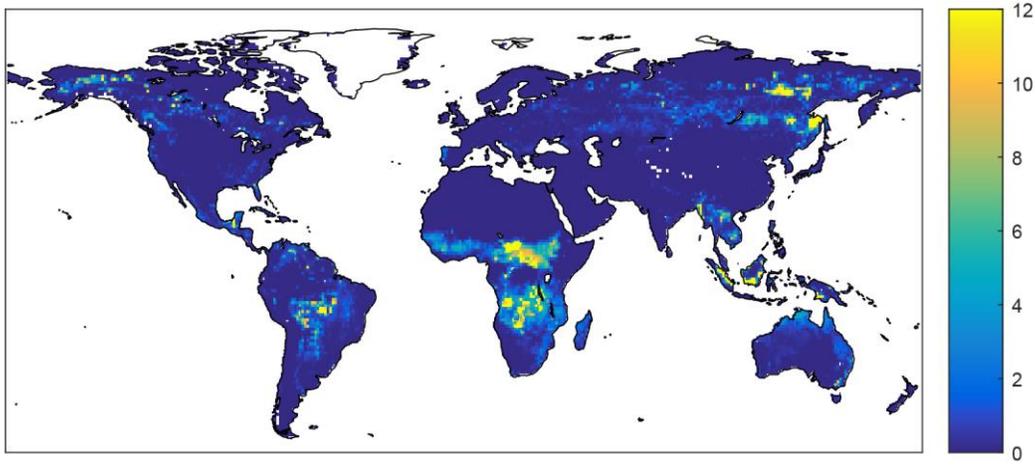
Figure S11. Fire N emissions (kg N ha^{-1} yr^{-1}) input in the atmospheric chemistry models that were used to derive the ACCMIP dataset of atmospheric N deposition. Note that the axis scale has been cutoff at 12 kg N ha^{-1} yr^{-1} for comparability with Figure S10a.
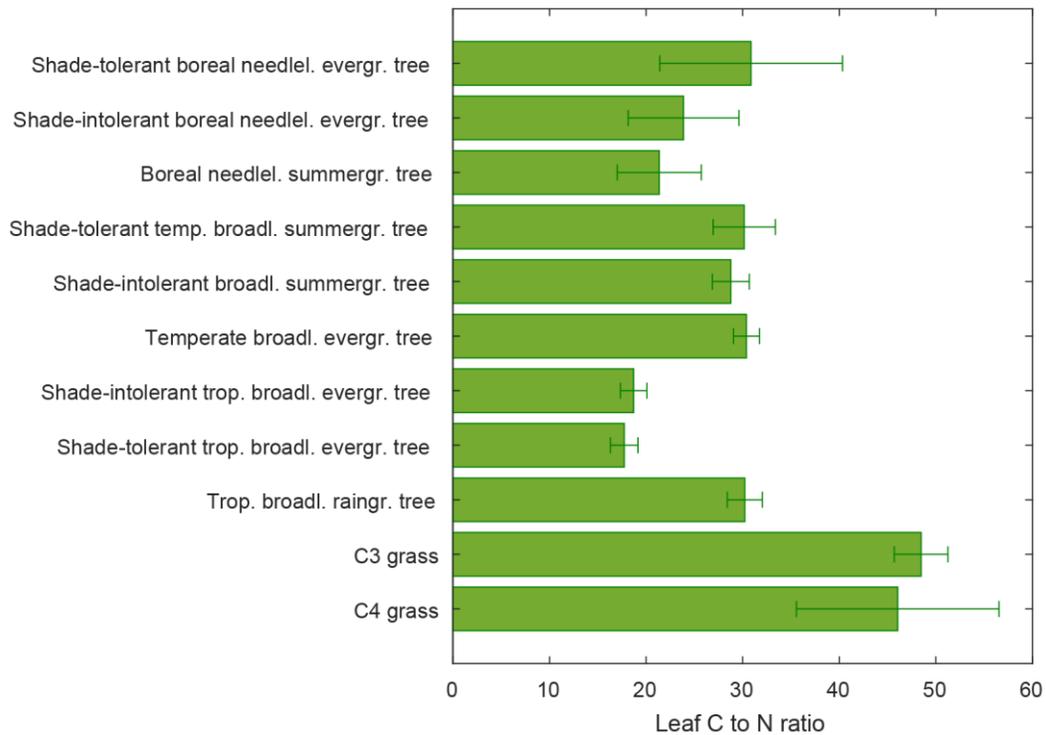
Figure S12. Predicted leaf C to N ratio of the plant functional types in LPJ-GUESS. The bars show means over the grid cells where the respective PFTs are dominant (have highest leaf area index). Error bars indicate 1 standard deviation.

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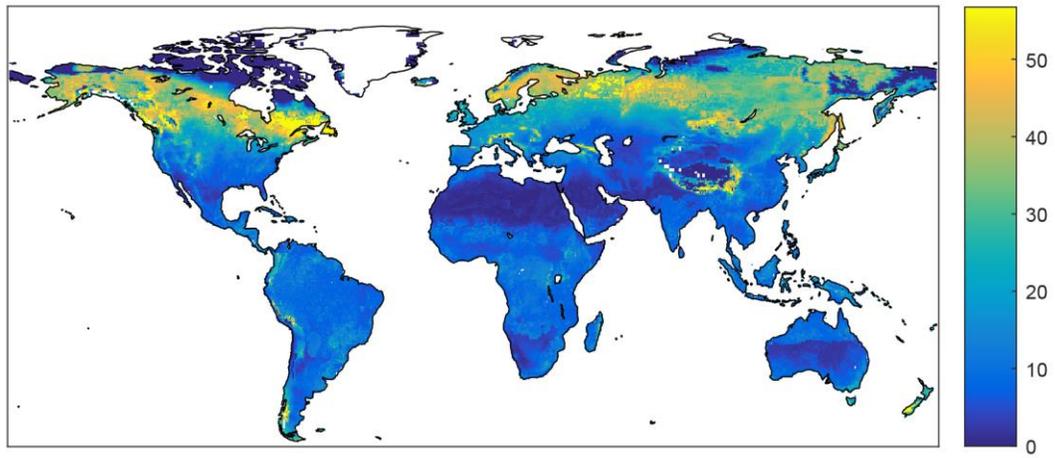
Figure S13. Soil organic carbon storage (kg C m^{-2}) for the true historical simulation (+Ndep +clim +CO$_2$) averaged over the period 1997–2006.
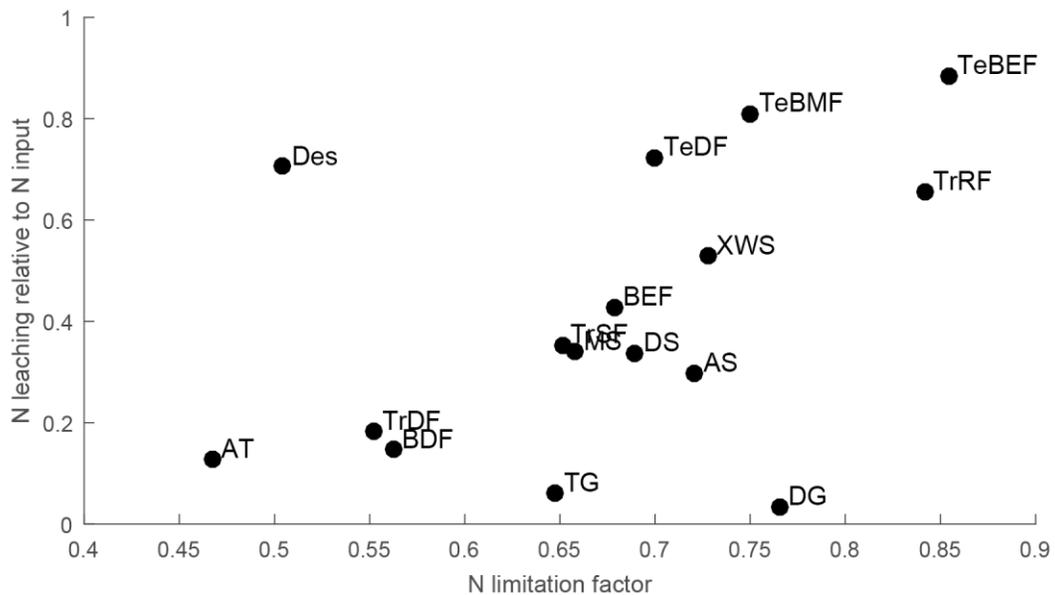
Figure S1411. N leaching relative to total N input vs N status averaged over the period 1997–2006 for a simulation run with true drivers (+Ndep +clim +CO$_2$) but fire and other disturbances switched off. TrRF: Tropical rainforest, TrDF: Tropical deciduous forest, TrSF: Tropical seasonal forest, BEF: Boreal evergreen forest/woodland, BDF: Boreal deciduous forest/woodland, TeBEF: Temperate broadleaved evergreen forest, TeDF: Temperate deciduous forest, TeBMF: Temperate/boreal mixed forest, TeMF: Temperate mixed forest, XWS: Xeric woodland/shrubland, MS: Moist savannah, DS: Dry savannah, AT: Arctic/alpine tundra, TG: Tall grassland, AS: Arid shrubland/steppe, DG: Dry grassland, Des: Desert.
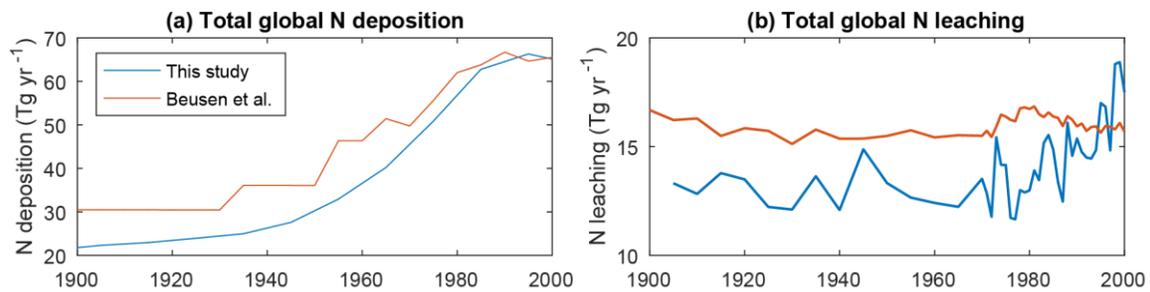
Figure S15~~12~~. Comparison of N fluxes with results of Beusen et al., (2016). (a) Total global N deposition, including non-natural lands. (b) Total global N deposition from natural lands, corrected for changes in natural landcover based on the dataset used by Beusen et al. (2016).
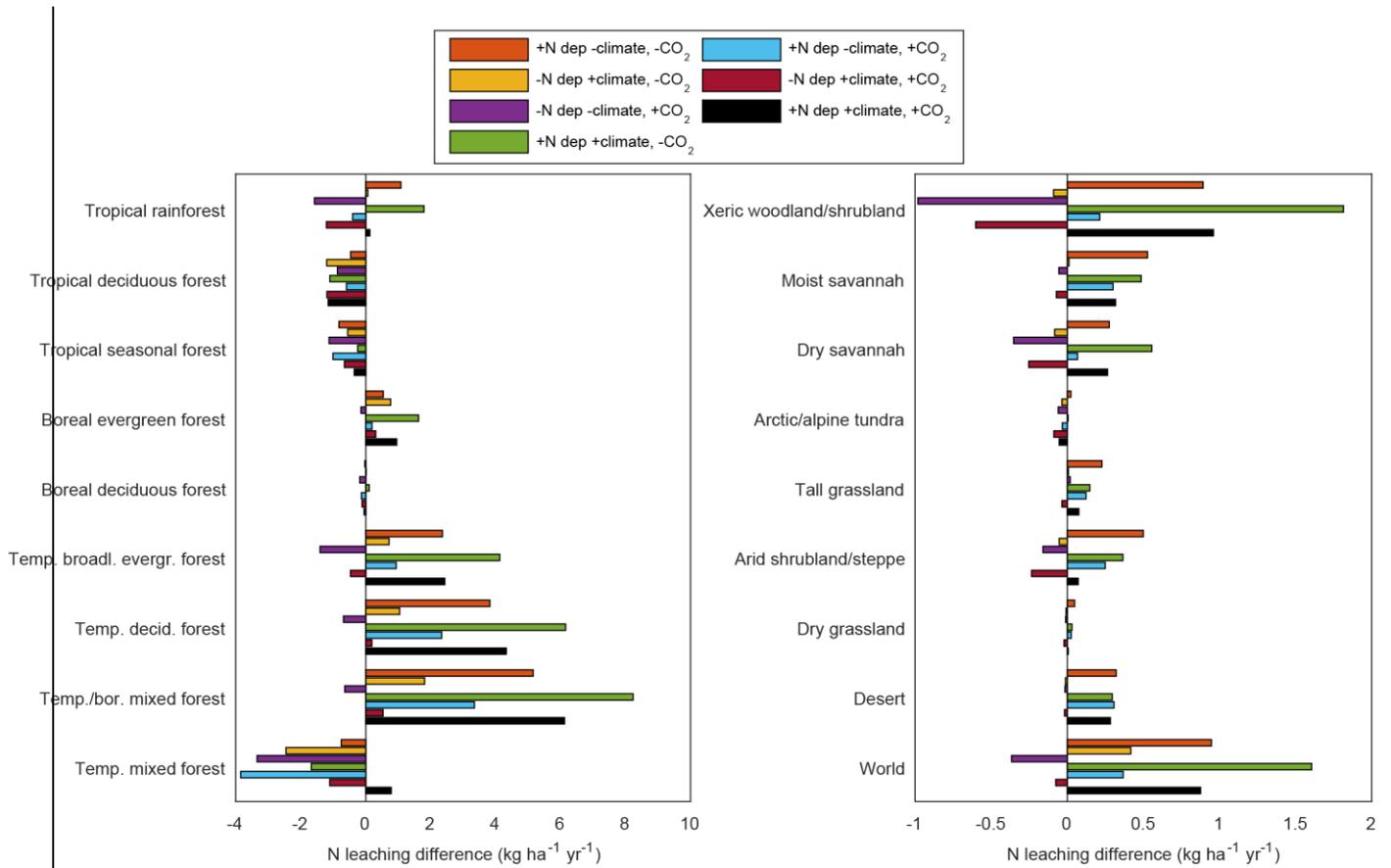
Figure S16~~13~~. N leaching difference with control simulation (-Ndep -clim -CO$_2$) for the eight simulations averaged over the period 1997–2006.

True N deposition, true climate, constant CO_2

True N deposition, constant climate, true CO_2

Constant N deposition, true climate, true CO_2
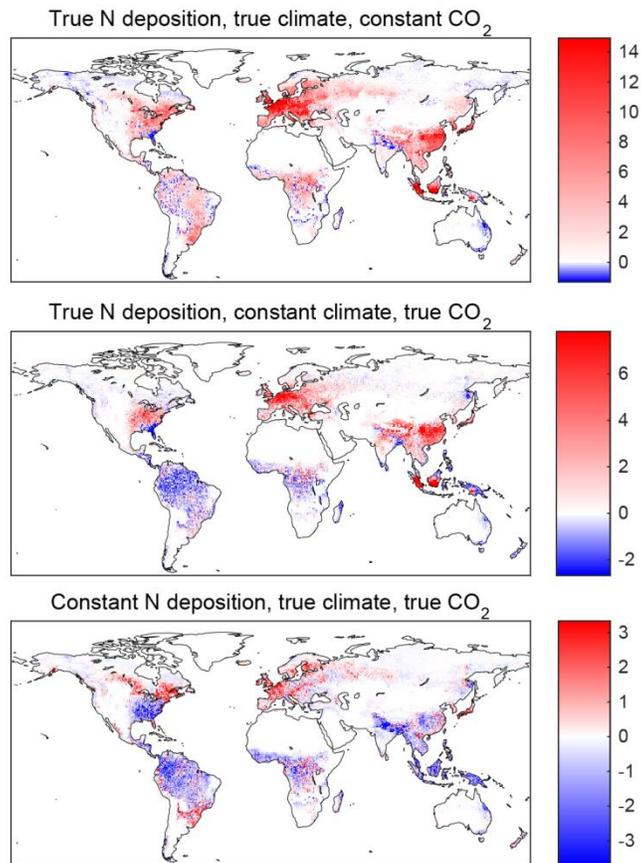
Figure S17~~14~~. Mineral N leaching difference with the control simulation (-Ndep -clim -CO$_2$) for the two-factor simulations. For ~~readability~~readability, the color axis has been cut off at approximately the 1% and 99% quantile.
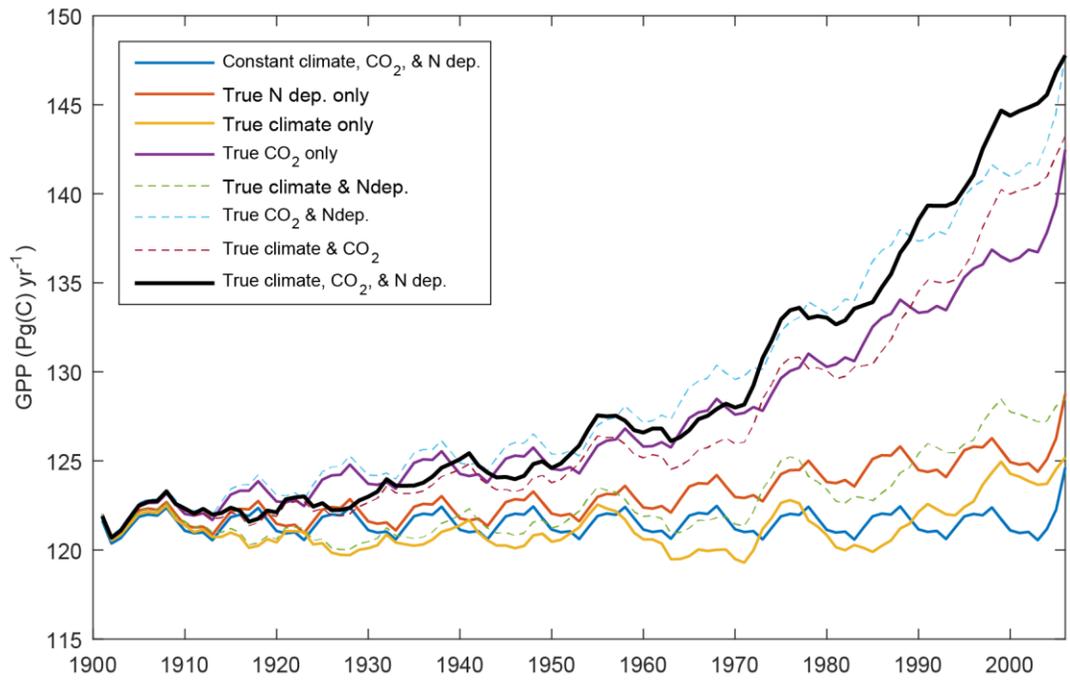
Figure S18~~15~~. Global total GPP vs time for the eight simulations. For ~~readability~~readability, the time series have been smoothed with a 5 year moving window
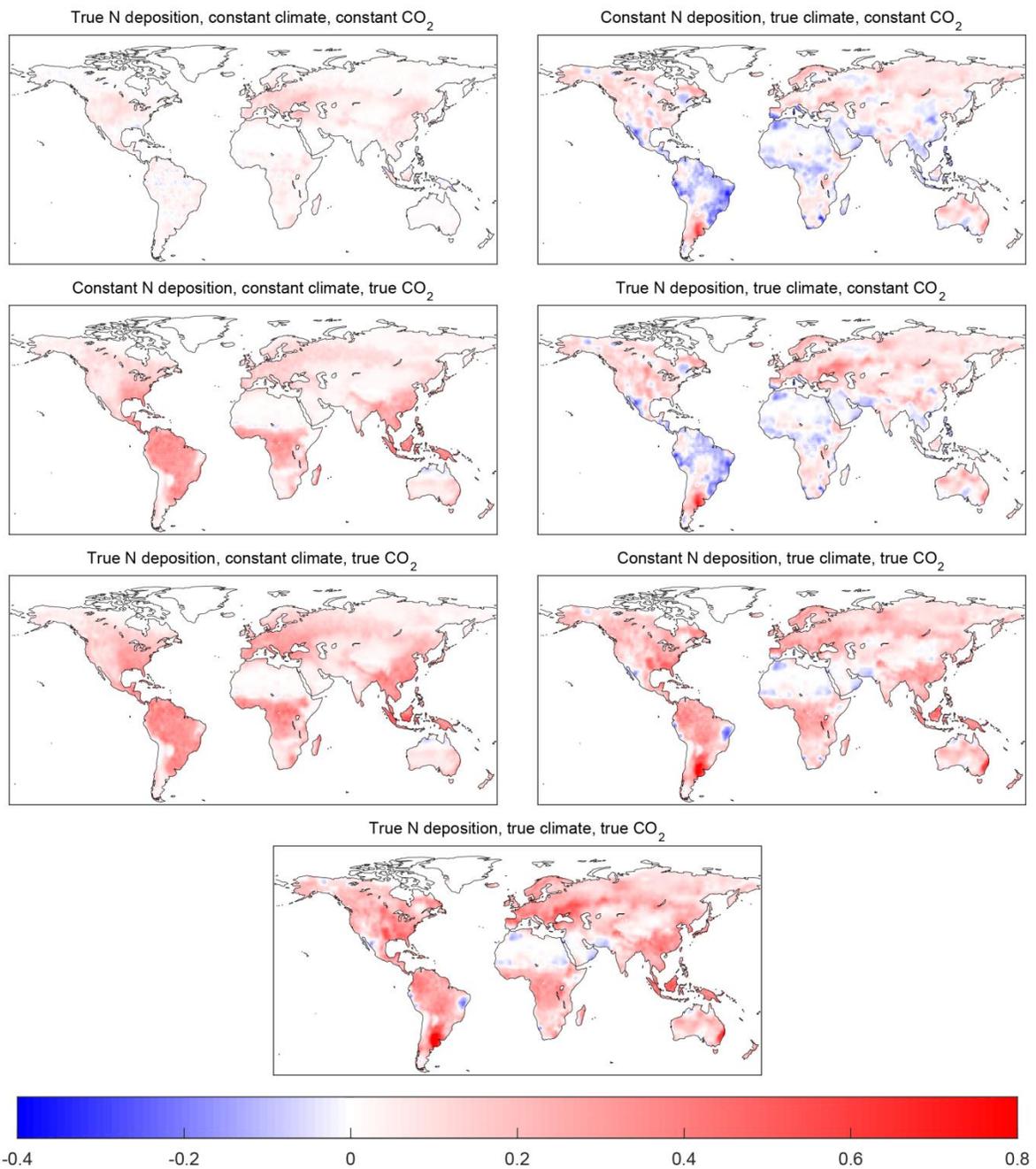
Figure S19~~16~~. GPP difference (kg-C m^{-2} yr^{-1}) with control simulation (-Ndep -clim -CO$_2$) for the other simulations averaged over the period 1997–2006.
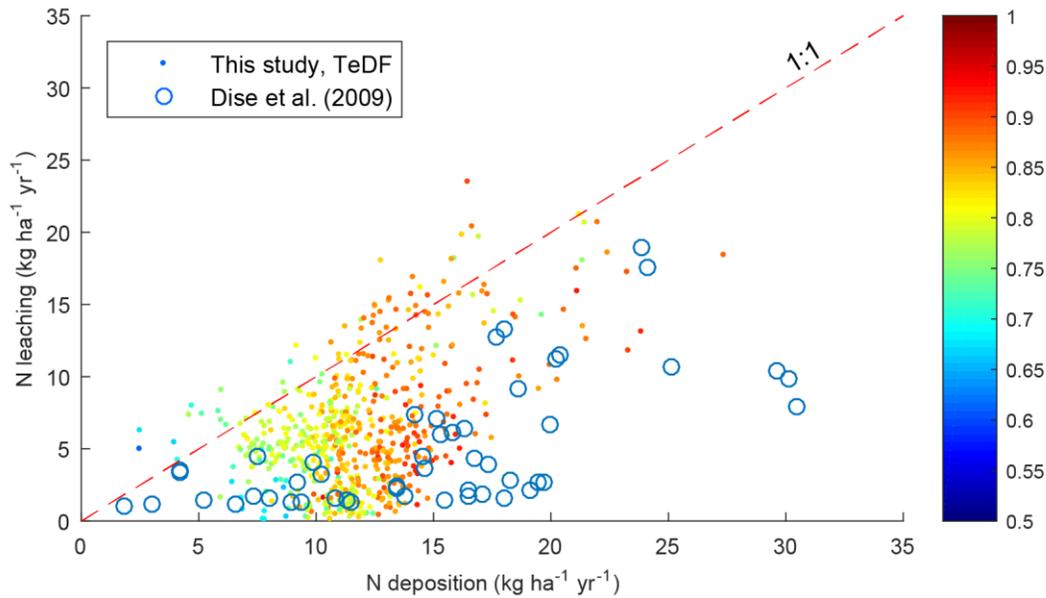
Figure S2017. N leaching vs N deposition in European temperate deciduous forests for the true historical simulation (+Ndep +clim +CO₂) (averaged over the period 1997–2006) and Level II sites of the UN-ECE/EC Intensive Monitoring Programme (Dise et al., 2009). Colors indicate ecosystem N status.
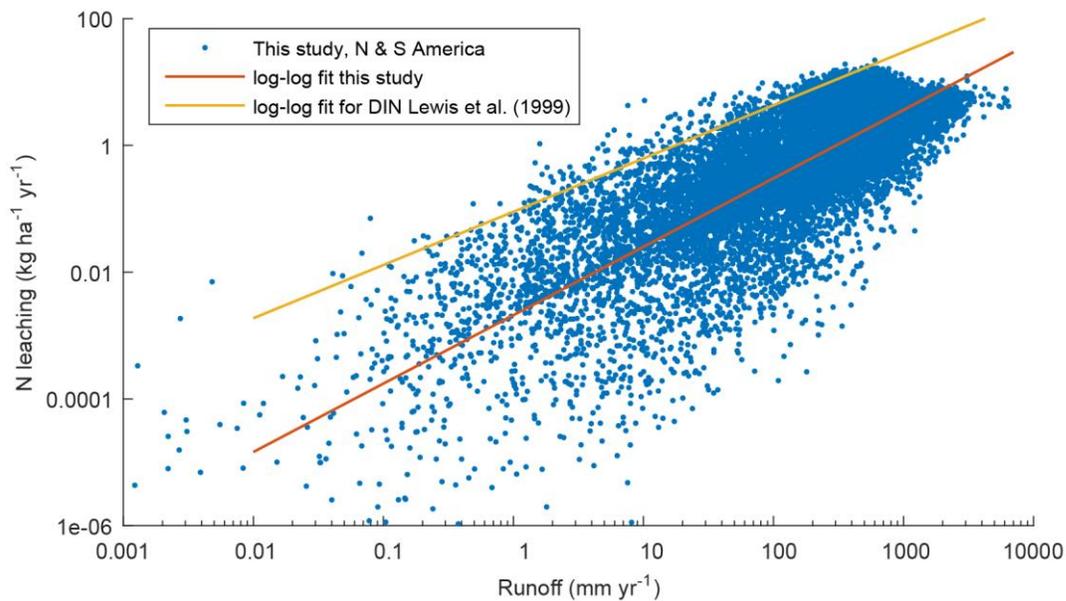
Figure S2118. Mineral N leaching vs runoff for North and South America. The linear fit on log-log scale is compared to a fit for dissolved inorganic nitrogen (DIN) losses vs runoff published by Lewis et al. (1999)Lewis et al., (1999).
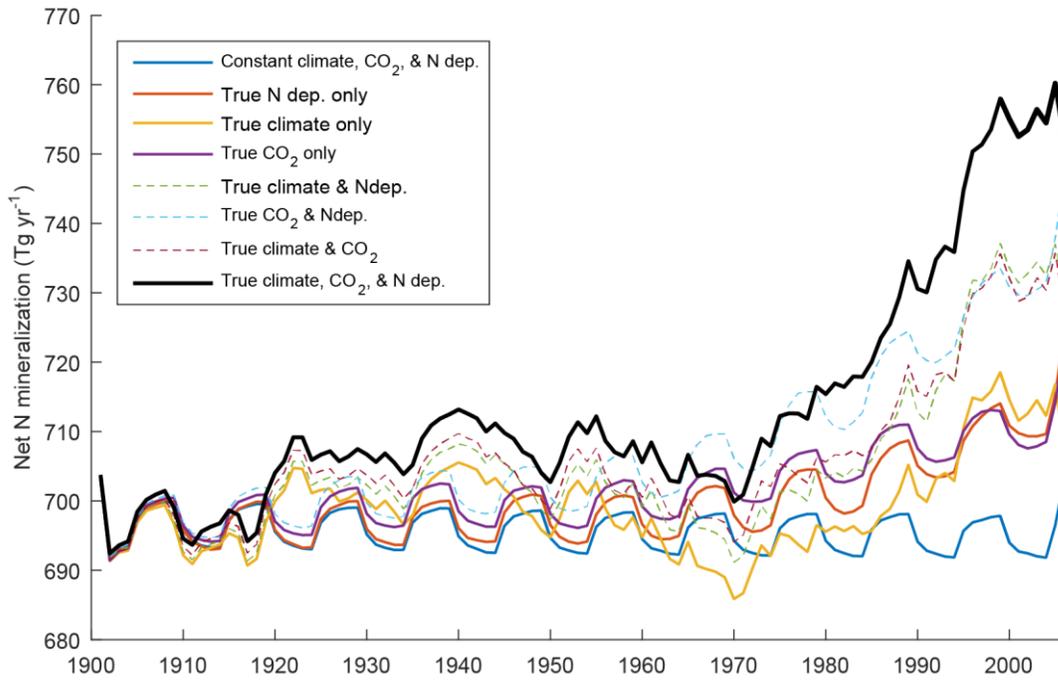
Figure S22~~19~~. Global total net N mineralization vs time for the eight simulations. For ~~readability~~readability, the time series have been smoothed with a 5 year moving window.

	TrRF	TrDF	TrSF	BEF	BDF	TeBEF	TeDF	TeBMF	TeMF	XWS	MS	DS	AT	TG	AS	DG	Des
TrRF	-	0.655	0.000	0.000	0.000	0.063	0.000	0.000	0.728	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TrDF	0.655	-	0.000	0.000	0.000	0.040	0.000	0.000	0.557	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TrSF	0.000	0.000	-	0.000	0.000	0.001	0.000	0.000	0.142	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BEF	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BDF	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeBEF	0.063	0.040	0.001	0.000	0.000	-	0.000	0.000	0.384	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeDF	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.047	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeBMF	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.043	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeMF	0.728	0.557	0.142	0.000	0.000	0.384	0.047	0.043	-	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000
XWS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.001	0.000	0.000	0.000
MS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000
DS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000
AT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000
TG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	-	0.000	0.000	0.000
AS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000
DG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000
Des	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-

Table S1. Test statistic of Welch's t-test for determining differences between biomes in total ecosystem N input (mean 1997–2006) between biomes (c.f. Fig. 2, main text). Welch's t-test is used for comparing populations with different variances with different and samples sizes. Significant differences ($\alpha \leq 0.05$) are printed in red. TrRF: Tropical rainforest, TrDF: Tropical deciduous forest, TrSF: Tropical seasonal forest, BEF: Boreal evergreen forest/woodland, BDF: Boreal deciduous forest/woodland, TeBEF: Temperate broadleaved evergreen forest, TeDF: Temperate deciduous forest, TeBMF: Temperate/boreal mixed forest, TeMF: Temperate mixed forest, XWS: Xeric woodland/shrubland, MS: Moist savannah, DS: Dry savannah, AT: Arctic/alpine tundra, TG: Tall grassland, AS: Arid shrubland/steppe, DG: Dry grassland, Des: Desert.

	TrRF	TrDF	TrSF	BEF	BDF	TeBEF	TeDF	TeBMF	TeMF	XWS	MS	DS	AT	TG	AS	DG	Des
TrRF	-	0.830	0.005	0.000	0.000	0.027	0.000	0.000	0.018	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000
TrDF	0.830	-	0.027	0.000	0.000	0.082	0.001	0.000	0.000	0.000	0.019	0.000	0.000	0.000	0.000	0.000	0.000
TrSF	0.005	0.027	-	0.000	0.000	0.000	0.192	0.000	0.000	0.000	0.442	0.000	0.000	0.000	0.000	0.000	0.000
BEF	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BDF	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeBEF	0.027	0.082	0.000	0.000	0.000	-	0.000	0.000	0.004	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeDF	0.000	0.001	0.192	0.000	0.000	0.000	-	0.000	0.000	0.000	0.011	0.000	0.000	0.000	0.000	0.000	0.000
TeBMF	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.476	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeMF	0.018	0.000	0.000	0.000	0.000	0.004	0.000	0.476	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
XWS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000
MS	0.002	0.019	0.442	0.000	0.000	0.000	0.011	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000
DS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000
AT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000
TG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000
AS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000
DG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000
Des	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-

Table S2. Test statistic of Welch's t-test for determining differences between biomes in total ecosystem N loss (mean 1997–2006) between biomes (c.f. Fig. 2, main text). Welch's t-test is used for comparing populations with different variances with different and samples sizes. Significant differences ($\alpha \leq 0.05$) are printed in red. TrRF: Tropical rainforest, TrDF: Tropical deciduous forest, TrSF: Tropical seasonal forest, BEF: Boreal evergreen forest/woodland, BDF: Boreal deciduous forest/woodland, TeBEF: Temperate broadleaved evergreen forest, TeDF: Temperate deciduous forest, TeBMF: Temperate/boreal mixed forest, TeMF: Temperate mixed forest, XWS: Xeric woodland/shrubland, MS: Moist savannah, DS: Dry savannah, AT: Arctic/alpine tundra, TG: Tall grassland, AS: Arid shrubland/steppe, DG: Dry grassland, Des: Desert.

	TrRF	TrDF	TrSF	BEF	BDF	TeBEF	TeDF	TeBMF	TeMF	XWS	MS	DS	AT	TG	AS	DG	Des
TrRF	-	0.231	0.106	0.000	0.374	0.000	0.000	0.383	0.000	0.950	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TrDF	0.231	-	0.034	0.889	0.012	0.000	0.000	0.127	0.000	0.331	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TrSF	0.106	0.034	-	0.000	0.329	0.001	0.000	0.528	0.000	0.378	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BEF	0.000	0.889	0.000	-	0.000	0.000	0.000	0.000	0.000	0.130	0.000	0.000	0.000	0.000	0.000	0.000	0.000
BDF	0.374	0.012	0.329	0.000	-	0.000	0.000	0.932	0.000	0.543	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeBEF	0.000	0.000	0.001	0.000	0.000	-	0.000	0.000	0.006	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeDF	0.000	0.000	0.000	0.000	0.000	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeBMF	0.383	0.127	0.528	0.000	0.932	0.000	0.000	-	0.001	0.693	0.000	0.000	0.000	0.000	0.000	0.000	0.000
TeMF	0.000	0.000	0.000	0.000	0.000	0.006	0.000	0.001	-	0.000	0.026	0.006	0.099	0.025	0.059	0.001	0.951
XWS	0.950	0.331	0.378	0.130	0.543	0.002	0.000	0.693	0.000	-	0.000	0.000	0.000	0.000	0.000	0.000	0.000
MS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.026	0.000	-	0.058	0.000	0.396	0.190	0.001	0.000
DS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.006	0.000	0.058	-	0.000	0.009	0.010	0.000	0.000
AT	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.099	0.000	0.000	0.000	-	0.000	0.002	0.000	0.000
TG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.025	0.000	0.396	0.009	0.000	-	0.404	0.114	0.000
AS	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.059	0.000	0.190	0.010	0.002	0.404	-	0.615	0.000
DG	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.001	0.000	0.000	0.114	0.615	-	0.000
Des	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.951	0.000	0.000	0.000	0.000	0.000	0.000	0.000	-

Table S3. Test statistic of Welch's t-test for determining differences between biomes in N net ecosystem exchange (mean 1997–2006) between biomes (c.f. Fig. 2, main text). Welch's t-test is used for comparing populations with different variances with different and samples sizes. Significant differences ($\alpha \leq 0.05$) are printed in red. TrRF: Tropical rainforest, TrDF: Tropical deciduous forest, TrSF: Tropical seasonal forest, BEF: Boreal evergreen forest/woodland, BDF: Boreal deciduous forest/woodland, TeBEF: Temperate broadleaved evergreen forest, TeDF: Temperate deciduous forest, TeBMF: Temperate/boreal mixed forest, TeMF: Temperate mixed forest, XWS: Xeric woodland/shrubland, MS: Moist savannah, DS: Dry savannah, AT: Arctic/alpine tundra, TG: Tall grassland, AS: Arid shrubland/steppe, DG: Dry grassland, Des: Desert.

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