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Bedrock Weathering Controls on Terrestrial Carbon-Nitrogen-Climate Interactions

Pawlok Dass[1,2], Benjamin Z. Houlton[1,3,4], Yingping Wang[5], David Wårlind[6], and Scott Morford[7]

[1]Department of Land, Air and Water Resources, University of California, Davis, Davis, CA, USA, [2]School of Informatics, Computing and Cyber Systems, Northern Arizona University, Flagstaff, AZ, USA, [3]John Muir Institute of the Environment, University of California, Davis, Davis, CA, USA, [4]Department of Global Development and Department of Ecology and Evolutionary Biology, Cornell University, Ithaca, NY, USA, [5]CSIRO Oceans and Atmosphere, Aspendale, Vic, Australia, [6]Department of Physical Geography and Ecosystem Science, Lund University, Lund, Sweden, [7]University of Montana, Missoula, MT, USA

Key Points:

- Bedrock nitrogen weathering contributes substantially to global carbon-nitrogen interactions
- Rock N inputs and biological N fixation contribute 2–5 times more to terrestrial C uptake than atmospheric N deposition though year 2101
- N deposition bypasses the C cycle and N inputs via weathering and fixation help to mitigate against progressive N limitations on the C sink

Supporting Information:

Supporting Information may be found in the online version of this article.

Correspondence to:

P. Dass,
pawlok.dass@gmail.com

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Author Contributions:

Conceptualization: Pawlok Dass, Benjamin Z. Houlton
Data curation: Pawlok Dass, Benjamin Z. Houlton
Formal analysis: Pawlok Dass, Benjamin Z. Houlton, Yingping Wang, David Wårlind, Scott Morford
Funding acquisition: Benjamin Z. Houlton
Investigation: Pawlok Dass, Benjamin Z. Houlton

Abstract

Anthropogenic nitrogen deposition is widely considered to increase CO_2 sequestration by land plants on a global scale. Here, we demonstrate that bedrock nitrogen weathering contributes significantly more to nitrogen-carbon interactions than anthropogenic nitrogen deposition. This working hypothesis is based on the introduction of empirical results into a global biogeochemical simulation model over the time period of the mid-1800s to the end of the 21st century. Our findings suggest that rock nitrogen inputs have contributed roughly 2–11 times more to plant CO_2 capture than nitrogen deposition inputs since pre-industrial times. Climate change projections based on RCP 8.5 show that rock nitrogen inputs and biological nitrogen fixation contribute 2–5 times more to terrestrial carbon uptake than anthropogenic nitrogen deposition though year 2101. Future responses of rock N inputs on plant CO_2 capture rates are more signficant at higher latitudes and in mountainous environments, where geological and climate factors promote higher rock weathering rates. The enhancement of plant CO_2 uptake via rock nitrogen weathering partially resolves nitrogen-carbon discrepancies in Earth system models and offers an alternative explanation for lack of progressive nitrogen limitation in the terrestrial biosphere. We conclude that natural N inputs impart major control over terrestrial CO_2 sequestration in Earth's ecosystems.

Plain Language Summary

Nitrogen is necessary for life; yet the availability of this nutrient is considered scarce when compared to plant demands in many of Earth's ecosystems. We show that bedrock weathering is an overlooked source of nitrogen that influences the terrestrial carbon cycle and plant carbon dioxide capture on a global scale. We find that the background sources of nitrogen prior to the industrial revolution, including bedrock sources and biological nitrogen fixation, have a substantial legacy effect on carbon dioxide removal that lasts hundreds of years. In contrast, modern increases in human-derived nitrogen deposition have a tendency to bypass the terrestrial carbon cycle, contributing to downstream water quality issues. We suggest that nitrogen constrains on plant carbon capture will be strongly influenced by rock weathering reactions in the 21st century, with implications for carbon-climate interactions and global warming.

1. Introduction

Carbon (C) and nitrogen (N) cycles are coupled from molecular to biosphere scales, with strong feedbacks on Earth's climate system (Galloway et al., 2004; Gruber & Galloway, 2008). Carbon sequestration, which is driven by net primary production (NPP), displays symptoms of N scarcity although 78% of the atmosphere is N (Vitousek & Howarth, 1991). Since bioavailable forms of N are mobile and readily lost from ecosystems, N inputs are needed to maintain C sinks (Cleveland et al., 2013). Consequently, an emerging view proposes that efforts aimed at reducing anthropogenic N deposition (originating primarily from agricultural and industrial activities) will reduce terrestrial C sinks, thereby obfuscating climate mitigation strategies (Gu et al., 2018; Hungate et al., 2003; Sutton et al., 2011; Thomas et al., 2010). However, chronic N deposition has been shown to cause harmful side-effects on terrestrial ecosystems, especially in Europe, North America and China, leading to N saturation (Aber et al., 1989; de Vries et al., 2014; Holland et al., 1997), resulting in reductions in terrestrial productivity and biodiversity (Aber et al., 1995), and degradation of aquatic

Methodology: Pawlok Dass, Benjamin Z. Houlton, Yingping Wang, David Wårlind, Scott Morford
Project Administration: Benjamin Z. Houlton
Resources: Pawlok Dass, Benjamin Z. Houlton
Software: Pawlok Dass, Benjamin Z. Houlton, Yingping Wang, David Wårlind
Supervision: Benjamin Z. Houlton
Validation: Pawlok Dass, Benjamin Z. Houlton
Visualization: Pawlok Dass, Benjamin Z. Houlton
Writing – original draft: Pawlok Dass, Benjamin Z. Houlton
Writing – review & editing: Pawlok Dass, Benjamin Z. Houlton, Yingping Wang, David Wårlind, Scott Morford

ecosystems downstream (Rabalais, 2002). Studies have revealed that between ∼10% and 23% of N fertilizers added to the soil are transported directly to groundwater and streams, respectively, without interacting with soil and plant pools, that is, completely bypassing or "short-circuiting" the terrestrial biosphere's C cycle (Schlesinger, 2009). This experimental finding has been further confirmed by natural stable isotope studies that have found small rain events to contribute large pulses of anthropogenic nitrogen losses to streamwaters (Wexler et al., 2014).

Conclusions drawn about the world's N balance and its interaction with the C cycle and climate system might also be sensitive to recent findings demonstrating the global-scale significance of rock N weathering in the terrestrial biosphere (Dahlgren, 1994; Houlton et al., 2018; Morford et al., 2011). Ecosystem models have highlighted the potential for progressive nitrogen limitations of plant productivity to emerge; yet studies have found mixed results and the terrestrial CO_2 sink continues to occur at ∼2 Gt C/yr, or ∼25% of global C emissions from human activities. Past studies have identified an important role for rock nitrogen weathering in nutrient cycles, ecological interactions, and regional carbon sequestration patterns across forests (Dynarski et al., 2019; Houlton & Morford, 2015; Morford et al., 2011), which could affect the nature and pattern of N limitation. Despite the evidence that rock weathering accounts for 26% of preindustrial N inputs, at the scale of the ecosystem, region and global, (Houlton et al., 2018), this pathway of N input has not been examined in global carbon-climate-nutrient models. Questions remain over the influence of rock N on the C cycle and how this pathway of N input might interact with other inputs in determining the degree of N saturation of soils and plants worldwide.

Here, we explore the full array of permutations and combinations of terrestrial N input pathways and their interactions with the global carbon cycle and climate change, including biological fixation, deposition, and rock N weathering. Through a set of global modeling experiments, we test the hypothesis that C sinks are more responsive to background (relatively older) N inputs via rock weathering (Houlton et al., 2018) and biological nitrogen fixation (Cleveland et al., 1999) (biological N fixation) than anthropogenic (relatively modern) N deposition (Lamarque et al., 2013). A corollary of this hypothesis is that, the modern anthropogenic N deposition will tend to saturate plant and soil N pools rather than stimulate CO_2 uptake (Aber et al., 1989) when background N inputs from rock weathering and biological fixation are explicitly considered in biogeochemical cycles (Methods Summary). We test this hypothesis both spatially across the terrestrial surface and temporally from the mid-1800s to 2101.

2. Materials and Methods

We conduct our geo-spatial analysis of C × N interactions via LPJ-GUESS (Smith et al., 2014), a widely used dynamic global vegetation model simulating both the C and N cycle. LPJ-GUESS was coupled with spatial simulations of N inputs via rock weathering (Houlton et al., 2018), biological N fixation (Cleveland et al., 1999) and atmospheric deposition (Lamarque et al., 2013), arriving at different points of time (on the geo-evolutionary timescale) across Earth's surface environment. Figure S1 illustrates the global distribution of the three N inputs for the pre-industrial period (mean for 1850–1879). All the inputs are assumed to interact with the terrestrial C cycle through the stoichiometric deficit of C/N interactions.

We focus our analysis on both the pre-industrial period (1850–1879) and future through year 2101. LPJ-GUESS captures empirically observed spatial patterns and magnitudes of NPP, with a global terrestrial NPP that falls within the range of other global C models (TRENDY) (Figures S4 and S5) (Sitch et al., 2015). Further, LPJ-GUESS considers a CO_2 fertilization effect consistent with free-air CO_2 enrichment experiments (Zaehle et al., 2014), thereby allowing for an analysis of terrestrial C x N x climate interactions.

Conceptually, we designed our modeling experiments to align with standard paradigms of changes in nutrient inputs during primary ecosystem succession. Although computational constraints prevent direct analysis of N inputs over geo-evolutionary time-scales in our model, we approximate background and historic C × N interactions, and quantify the effect of systematically altering the order of N inputs on biogeochemical cycles across the globe and for the entire terrestrial biosphere. We created a factorial matrix approach that explores all N input permutations and combinations and provides an uncertainty range that is, based on order of introduction into the model. Prior to large-scale human influences on the N cycle (beginning in the late 1800s, the beginning of the industrial revolution) ecosystems relied principally on

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the N inputs via relatively modest amounts of atmospheric deposition (primarily N fixed during lightning strikes) (Lelieveld & Dentener, 2000) and the more substantial N inputs via parent material weathering (Houlton et al., 2018) in addition to biological colonization of ecosystem pools that is, biological N fixation (Cleveland et al., 1999). Thus, in our model experiments, we first assume a background atmospheric N deposition flux via lightning strikes for all the cases studied in this paper to account for legacy N inputs since the beginning of Earth history. Rock weathering reactions accelerate substantially once lithified sediments are tectonically uplifted and exposed to Earth's surface environment. We thus allow for rock N inputs (Houlton et al., 2018) to occur second in our model, given the formation of such sediments dating back 100s of millions of years. Terrestrial Biological N fixation (Cleveland et al., 1999) inputs, which evolved toward a large scale symbiotic pathway around 60 million years ago (Sprent & Raven, 1985), is introduced after these N inputs in the pre-industrial analysis. While the reality is that small but important fluxes of biological N fixation can and do occur on un-weathered rocks, the more substantial biological N fixation inputs in litter and soil pools, and via symbiotic N fixation, occur after rock weathering reactions and soil formation processes are sufficiently well developed—and so our approach is also consistent with ecosystem development (Vitousek, 2004). In addition, as wildfires and early agrarian activities began influencing the N cycle (early to mid-1800s), we introduce the influence of such human actions on atmospheric N deposition (Lamarque et al., 2013) as the final N input after N from lightning, weathering (B. Z. Houlton et al., 2018) and biological N fixation (Cleveland et al., 1999). During the 20th and 21st centuries however, with widespread burning of fossil fuels, N deposition (Lamarque et al., 2013) increased substantially, adding to these background N inputs. Nonetheless, for the pre-industrial period, in order to disentangle the impact of the sequence of N inputs on NPP and N saturation, we conduct a full factorial analysis that considers all its permutations and combinations.

Specifically, we analyze the following set of scenarios in the pre-industrial period and in the following order:

1. LightningN (atmospheric N deposition originating only from lightning strikes)
2. RockN (N inputs only from rock weathering)
3. Nfix (N inputs only from biological N fixation)
4. NDEP (only atmospheric deposition of N from early agriculture)
5. RockN + Nfix (combination of 2 & 3 only)
6. Nfix + NDEP (combination of 3 & 4 only)
7. RockN + NDEP (combination of 2 & 4 only)
8. RockN + Nfix + NDEP (combination of 2, 3 & 4 only)

We then subtract scenarios from one another to quantify effects on NPP and N saturation:

$$\text{RockN}\left(1^{\text{st}}\right) = \text{RockN} - \text{LightningN} \tag{1a}$$

$$\text{RockN}\left(2^{\text{nd}}\right) = \left[\text{RockN} + \text{Nfix}\right] - \text{Nfix} \tag{1b}$$

$$\text{RockN}\left(3^{\text{rd}}\right) = \left[\text{RockN} + \text{Nfix} + \text{NDEP}\right] - \left[\text{Nfix} + \text{NDEP}\right] \tag{1c}$$

$$\text{Nfix}\left(1^{\text{st}}\right) = \text{Nfix} - \text{LightningN} \tag{2a}$$

$$\text{Nfix}\left(2^{\text{nd}}\right) = \left[\text{RockN} + \text{Nfix}\right] - \text{RockN} \tag{2b}$$

$$\text{Nfix}\left(3^{\text{rd}}\right) = \left[\text{RockN} + \text{Nfix} + \text{NDEP}\right] - \left[\text{RockN} + \text{NDEP}\right] \tag{2c}$$

$$\text{NDEP}\left(1^{\text{st}}\right) = \text{NDEP} - \text{LightningN} \tag{3a}$$

$$\text{NDEP}\left(2^{\text{nd}}\right) = \left[\text{Nfix} + \text{NDEP}\right] - \text{Nfix} \tag{3b}$$

$$\text{NDEP}\left(3^{\text{rd}}\right) = \left[\text{RockN} + \text{Nfix} + \text{NDEP}\right] - \left[\text{RockN} + \text{Nfix}\right] \tag{3c}$$

For our 20th and 21st century experiments, while rock N inputs (Houlton et al., 2018) change negligibly (Figures S6 and S7a) and biological N fixation (Cleveland et al., 1999) increases slightly (Figures S6 and S7b), atmospheric N deposition (Lamarque et al., 2013) shows a three-fold increase largely in response to fossil fuel combustion (Figures S6 and S7c). Thus, we further simplify our analysis by contrasting the impacts of post-industrial increases in N deposition with a combination of rock N and biological N fixation inputs, referred to as "background N inputs".

We used LPJ-GUESS (Smith et al., 2014) version 3.1, a dynamic global vegetation model (DGVM) which combines individual and patch based representation of vegetation dynamics with ecosystem biogeochemical cycling, in our spatial and global analyses. This DGVM includes explicit C × N interactions between plant, vegetation and soil, making it an appropriate model for investigating nutrient limitations on the terrestrial pattern and process, specifically vegetation C sinks, on a global basis. LPJ-GUESS includes 12 plant functional types (PFTs) whose parameters are generalized to approximate global mean conditions and analyzed in spatial grid-cells over time. It has a pool of mineral N available for plant root uptake and to soil microbes. The N inputs from rock weathering, biological fixation and atmospheric deposition contribute to this pool. There are 11 soil pools and four vegetation pools with differing C:N stochiometry. The dynamics of these pools determine the N demand. N limitation occurs when the N supply is less than the demand. The model performance was validated against both global totals and spatial patterns of GPP vis-à-vis empirical data and multi-model means, showing relatively good correspondence (Figures S4 and S5).

LPJ-GUESS was run at a spatial resolution of 0.5°Latitude/Longitude globally. All simulations were initialized using a 500-year spin-up cycle to build vegetation and soil C & N pools to a steady state. The spin-up used the first 30 years of climate forcing data (Jones & Harris, 2013) and CO_2 concentrations (RCP 8.5 scenario [Riahi et al., 2011]), that is, data from 1850 to 1879 for the pre-industrial simulation and 1901–1930 for the 20th and 21st century simulations. To initiate vegetation growth, the first 100 years of model spin-up were considered to be free of N constraints on NPP. For the pre-industrial conditions, simulations were carried out from 1850 to 1879 and the 30-year mean was presented. For the future climate change scenario, simulations were carried out from 1901 to 2101.

LPJ-GUESS does not include prescribed terrain data and thus depends on climate forcing data to simulate spatial heterogeneity of PFTs. We used climate data from Climatic Research Unit (CRU) (Jones & Harris, 2013) which is based on observations of spatial resolution at 0.5°latitude and longitude. For future projections, we used climate trends of the GCM (general circulation model) FGOALS-g2 (Li et al., 2013) for the RCP 8.5 (Riahi et al., 2011). Atmospheric CO_2 concentrations and atmospheric deposition of N for RCP 8.5 were also used in the model. We choose the RCP 8.5 scenario as it represents the 95th percentile of no policy coordination on GHG emissions and largely tracks with current emissions (Riahi et al., 2011).

Monthly atmospheric N-deposition rates were taken from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) database (Lamarque et al., 2013). The original data at 1.9° × 2.5°grid was interpolated to the climate grid of 0.5° × 0.5°. LPJ-GUESS was forced with global terrestrial N inputs via rock weathering (Houlton et al., 2018), biological N fixation (Cleveland et al., 1999) and atmospheric deposition (Lamarque et al., 2013). First, the N input flux from bedrock weathering was computed using a data-driven probabilistic model that incorporates topographic, climatological, and lithological factors to estimate N denudation and chemical weathering rates and calibrated using solute sodium (Na+) fluxes from large river basins across the globe (Houlton et al., 2018). The original model output at 1 km^2 scale was regridded and upscaled to match 0.5° × 0.5° grid of LPJ-GUESS. Second, biological N fixation is computed as a function of evapotranspiration that builds on an empirical correlation between N fixation and evapotranspiration at the global scale. We used the "conservative" parameters of the linear regression equation for predicting N fixation values (Cleveland et al., 1999) as based on actual evapotranspiration data (AET). Observed AET data were taken from MODIS-MOD16 (Mu et al., 2007) and future trends were taken

Table 1
Full Factorial Analysis of the Impact of Order of N Input Pathways on the Net Primary Production (ΔNPP) (Global Total). "1st, 2nd & 3rd" Implies the Respective Order in Which Each N Input Enters the Terrestrial Biosphere

ΔNPP (PgC/yr)	Rock N	Biological N fixation	N deposition
1st	**4.56**[Equation 1a]	10.88[Equation 2a]	7.36[Equation 3a]
2nd	0.36[Equation 1b]	**6.68**[Equation 2b]	0.57[Equation 3b]
3rd	0.20[Equation 1c]	2.72[Equation 2c]	**0.41**[Equation 3c]

Note. The labels in square brackets explicitly link to the respective model scenario equations in the text. Numbers in bold reflect our best estimate of the most conceptually plausible order in which N inputs enter ecosystems (locally and globally) as described in the text. These values are for the pre-industrial period, that is, mean of 1850–1879.

from GCM FGOALS-g2 (Li et al., 2013). Third, atmospheric deposition of N was prescribed from an external RCP scenario dependent data set (Lamarque et al., 2013). N inputs from all three sources are added to a common pool of mineral N. Future trends for all the N input responses to climate and CO2 change were computed using the RCP 8.5 scenario.

For the pre-industrial period, the global totals for N inputs (mean for 1850–1879) from rock weathering, biological N fixation and atmospheric deposition were equal to 15.5, 71.0 and 5 + 15.6 Tg N/yr (lightning plus early agrarian effects on N deposition (Galloway et al., 2004]), respectively. Spatially, rock N inputs (Figure S1a) were highest in mountainous ecosystems underlain by N-rich lithologies and among moisture climates where weathering rates peak (Houlton et al., 2018). The N fixation map (Figure S1b) captures the decrease in biological N fixation from the tropics to the sub-tropics, consistent with theory and empirical observations (Houlton et al., 2008). Although the global total for Nfix is larger than RockN, this analysis reveals the increasing relative importance of RockN inputs with increasing latitude. Moreover, while the global totals for RockN and NDEP appear similar, they differ in their spatial distributions. N deposition from lightning (~5 TgN/yr) was distributed globally and uniformly while the pre-industrial anthropogenic influence on N deposition (15.6 TgN/yr) was limited to agricultural belts and high human density centers (Figure S1c). Post-industrial N deposition (Lamarque et al., 2013) inputs consider growth in both fossil fuel combustion and synthetic fertilizer effects on atmospheric N emissions (Galloway et al., 2004).

3. Results and Discussion

Results of our global modeling analysis confirm the hypothesis that N inputs from rock weathering (Houlton et al., 2018), as a legacy N input that pre-dates human activities, exerts hitherto unrecognized leverage over plant CO_2 uptake and N saturation of ecosystem pools worldwide. This N input is shown to contribute significantly to the background N pools in plants and soils, which, by consequence, allows for atmospheric N deposition to "overtop" ecosystem pools and demands, leading to a by-pass of the C cycle. Over the pre-industrial baseline period (1850–1879), N inputs via rock weathering (Equation 1a) (Houlton et al., 2018) and biological N fixation (Equation 2b) (Cleveland et al., 1999) account for a change in NPP (ΔNPP) equal to 4.56 and 6.68 PgC/yr, which underlie global terrestrial NPP totals equal to 34.4 and 41.2 PgC/yr, respectively. In contrast, the contribution of N deposition (Equation 3c) (Lamarque et al., 2013) to NPP is an order of magnitude smaller (i.e., ΔNPP = 0.41 PgC/yr). This result is observed despite lower N inputs via rocks cf. deposition, pointing to the importance of background (pre-historic) contributions of weathering to vegetation and soil N pools and cycling compared to the increase in N deposition due to early agrarian activities (Table 1). For example, examining N deposition (Lamarque et al., 2013) in the absence of rock N (Houlton et al., 2018) inputs in our model, as all previous analyses have done (i.e., N deposition 1st scenario or [Equation 3a]), inflates N deposition's impact on ΔNPP to 7.36 PgC/yr (Table 1 right column; Methods Summary).

Spatially, ΔNPP ascribed to rock N weathering (Equation 1a) (Houlton et al., 2018) during the pre-industrial period (Figure S2a) is higher in the tropics than extra-tropics. Although rock N inputs are on average lower in tropical than temperate and boreal environments (Houlton et al., 2018), the long growing season, warm temperatures, and high annual rainfall rates underpin high photosynthetic demand for nutrients in tropical forests. This translates to a more substantial NPP response to a given quantity of N input. ΔNPP attributed to biological N fixation (Equation 2b) (Figure S2b) follows a similar pattern, but with an overall greater effect size, given the dominance of biological N fixation inputs in the tropics (Cleveland et al., 1999). These results do not imply that other nutrients, such as P, are less limiting than N in tropical environments; rather, NPP is poised to respond to N inputs where conditions and resources for photosynthesis are maximized. The role of atmospheric N deposition (Equation 3c) (Figure S2c) in spatial NPP patterns is considerably smaller than for either rock N or biological N fixation inputs; its smaller pre-industrial (vs. modern) magnitude, coupled with lack of significant contribution to tropical areas, reduces N deposition's contribution to NPP in the pre-industrial era.

Table 2

Global ΔNPP: ΔNleach (in PgC:TgN) Ratio for the Pre-Industrial Baseline Period (Mean of 1850–1879)

Order of introduction in model	Rock N ΔNPP:ΔNleach (PgC:TgN)	Biological N fixation ΔNPP:ΔNleach (PgC:TgN)	N deposition ΔNPP:ΔNleach (PgC:TgN)
1st	**0.63** [Equation 1a]	0.25 [Equation 2a]	1.04 [Equation 3a]
2nd	0.03 [Equation 1b]	**0.14** [Equation 2b]	0.03 [Equation 3b]
3rd	0.02 [Equation 1c]	0.05 [Equation 2c]	**0.02** [Equation 3c]

Note. This factorial matrix approach allows the exploration of all permutations and combinations of order of all three N inputs. The labels in square brackets explicitly link to the respective equations of N inputs.

To further analyze the environmental benefit (C uptake) (Table 1) versus risks (N losses) (Table S1) of individual N input pathways, we develop a quantitative index, ΔNPP:ΔNleach (Table 2), where ΔNleach denotes the change in N leaching loss. We show that the ΔNPP:ΔNleach of a given N input pathway decreases in proportion to its order of appearance in our model simulations; that is, the earlier a given N input enters terrestrial nutrient cycles, the greater its impact on C sequestration versus N saturation (Tables 1, 2 and S1; Materials & Methods). Consequently, rock N weathering (Equation 1a) and biological N fixation (Equation 2b) have a larger influence on terrestrial NPP, resulting in strong biological retention of these N sources, whereas the more contemporary rise in atmospheric N deposition (Equation 3c) via human activities disproportionately accelerates losses of N from the land compared to its effect on NPP. This finding makes logical sense, especially from the point of view of a "tipping bucket model": background or older N inputs via weathering and biological fixation satisfy terrestrial N demands, with additional N inputs progressively "overtopping" the capacity for nutrient retention, thereby short-circuiting the C cycle. Moreover, under pre-industrial conditions, rock weathering (Equation 1a) is shown to be the most efficient N input pathway, storing 0.63 PgC/TgN lost to the environment (Figure S3). Atmospheric N deposition (Equation 3c), at the other extreme, is 30 times less efficient in promoting C uptake (Figure S3) (Table 2).

We address the impact of 21st century climate changes and GHG emissions on our results by examining IPCC's RCP8.5 scenario (Riahi et al., 2011) (projected to cause a temperature increase of ~2.6°C–4.8°C [Collins et al., 2013]), where atmospheric CO_2 concentrations and N deposition increase substantially through 2101 (Figure S6) due to a large increases of fossil fuel burning. We also consider the climate-dependence (Houlton et al., 2008) of N fixation in such simulations (Figure S6). While direct effects of N inputs on CO_2 fertilization (Baker & Boote, 1996) and indirect effects of climate change on terrestrial NPP are well examined (Ciais et al., 2014), the collective ensemble of N deposition impacts on C sinks remains controversial on account of the large uncertainty around the fate of the majority (65%) of the N inputs to the terrestrial biosphere (Galloway et al., 2008).

Despite increased anthropogenic N deposition and atmospheric CO_2 concentrations, our prognostic model reveals greater efficiency of C sequestration via legacy pre-anthropogenic N input pathways (rock N + biological N fixation), that is, higher ΔNPP:ΔNleach ratios (Figure 1c, Figure 3a), than modern increases in atmospheric N deposition via human activities through 2101. Spatially, the pre-anthropogenic background N inputs result in the highest NPP increases in the tropics and higher latitudes (Figure 2a1). Rock N supports NPP increases in mountainous areas with substantial tectonic uplift in addition to climates where precipitation exceeds evapotranspiration (Houlton et al., 2018) (Figure S9a1) while biological N fixation, similar to the pre-industrial results, supports the largest positive trends in NPP in the tropics (Figure S9b1). In terms of temporal changes, background N inputs underlie predominantly positive ΔNPP:ΔNleach trends worldwide (Figure 3a). Over the period of 1901–2101, C gains expand relative to N losses among biological N fixation and rock N inputs. Contemporary increases in atmospheric N deposition driven largely by fossil fuel combustion, in contrast, disproportionately accelerate N losses over the same time period. Accordingly, our model simulates uniformly low ΔNPP:ΔNleach (Figure 1c) (N losses outweigh C gains) and predominantly negative temporal trends in this ratio, especially in the mid- to high-latitudes in response to rising N deposition (Figure 3b). Collectively, these results highlight the fundamental importance of background N cycle processes in determining C cycle and climate impacts of N management, both now and into the future.

3.1. Uncertainties and Future Work

A set of indirect observations support our model-based findings, though there are important uncertainties to consider. Our model predicts ~72 kg C sequestration/kg N deposition in contemporary ecosystems, with a marginal increase of ~85 kg C/kgN simulated through year 2101 (Figure S11). These estimates are consistent with the range of 51–82 kg vegetation C stored/kg N input observed for forest inventory analysis (Thomas et al., 2010). In addition, the fraction of N deposition incorporated into terrestrial vegetation rises

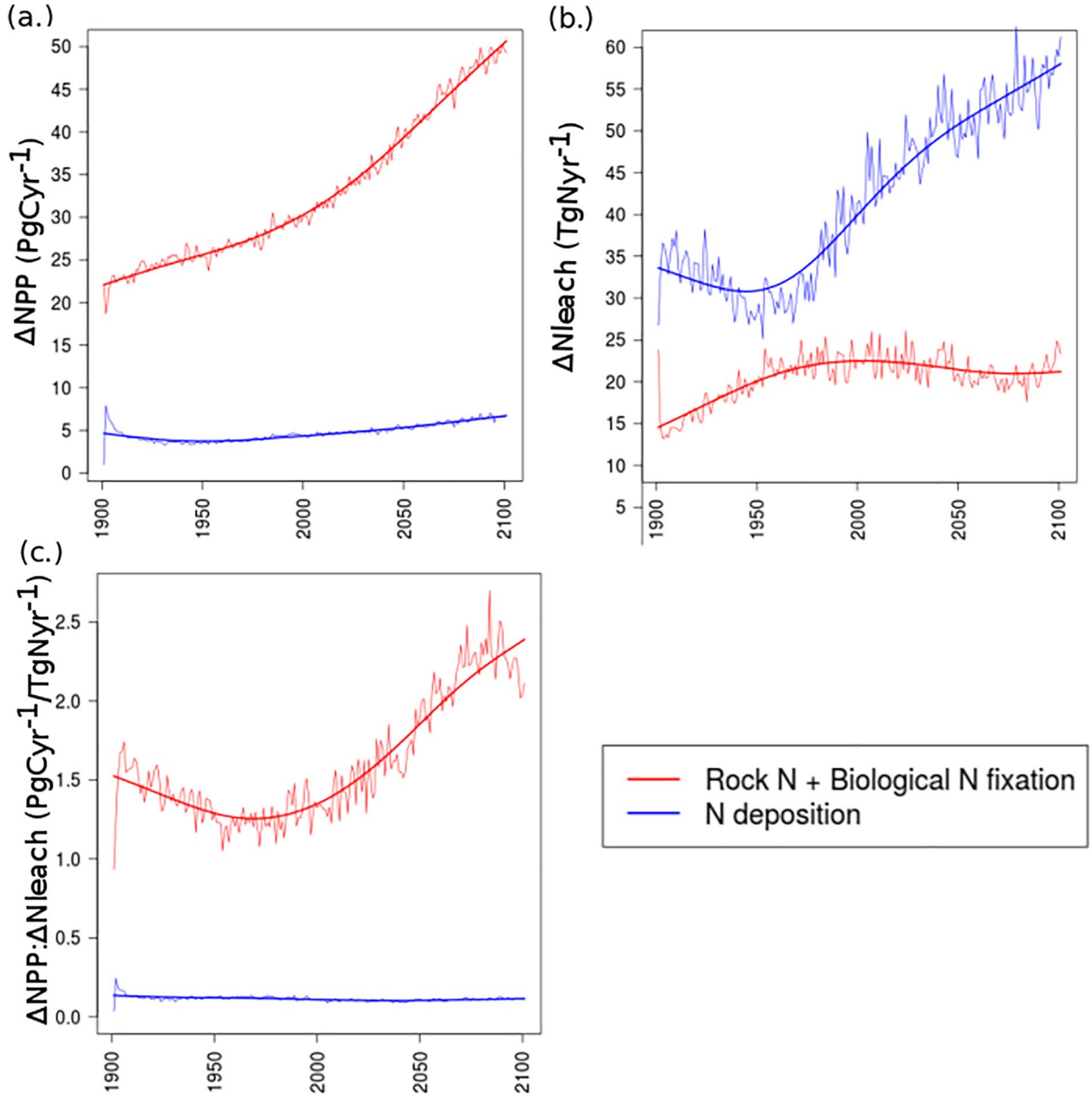

Figure 1. Annual (a) net primary production (ΔNPP) (PgC), (b) ΔNleach (TgN) and (c) ratio of ΔNPP to ΔNleach for background N inputs (rock N + biological N fixation) and N deposition. A smoothing spline with a spar value of 1 was used. For individual N inputs, that is, rock N and biological N fixation separately, see Figure S8.

from a pre-industrial value of 5% to a present day (2000–2010 mean) value of ~10% in our global model. A literature analysis using ^{15}N experiments suggested that between 0% and 40% of N fertilizer additions are incorporated into plants (Schlesinger, 2009). In reality, N leaching losses are likely higher than computed here, since LPJ-GUESS has been found to underestimate the N leaching flux by ~ 4 times of observed values (Smith et al., 2014). From a global modeling perspective, our calculations suggest that the magnitude of NPP attributed to rock N inputs might resolve some of the extant N cycle related gaps in the residual land carbon sink and the land carbon uptake simulated by Earth system models (Ciais et al., 2014; Hungate et al., 2003). However, while past studies (Moulton et al., 2000) suggest that plants and microbes can increase weathering rates by a factor of 2–10, we did not consider such geo-biological effects in our model, implying that our estimates for N inputs from rock weathering might be conservative (Houlton et al., 2018). Moreover, other elements, such as P, are known to influence the terrestrial C cycle, with interactions with

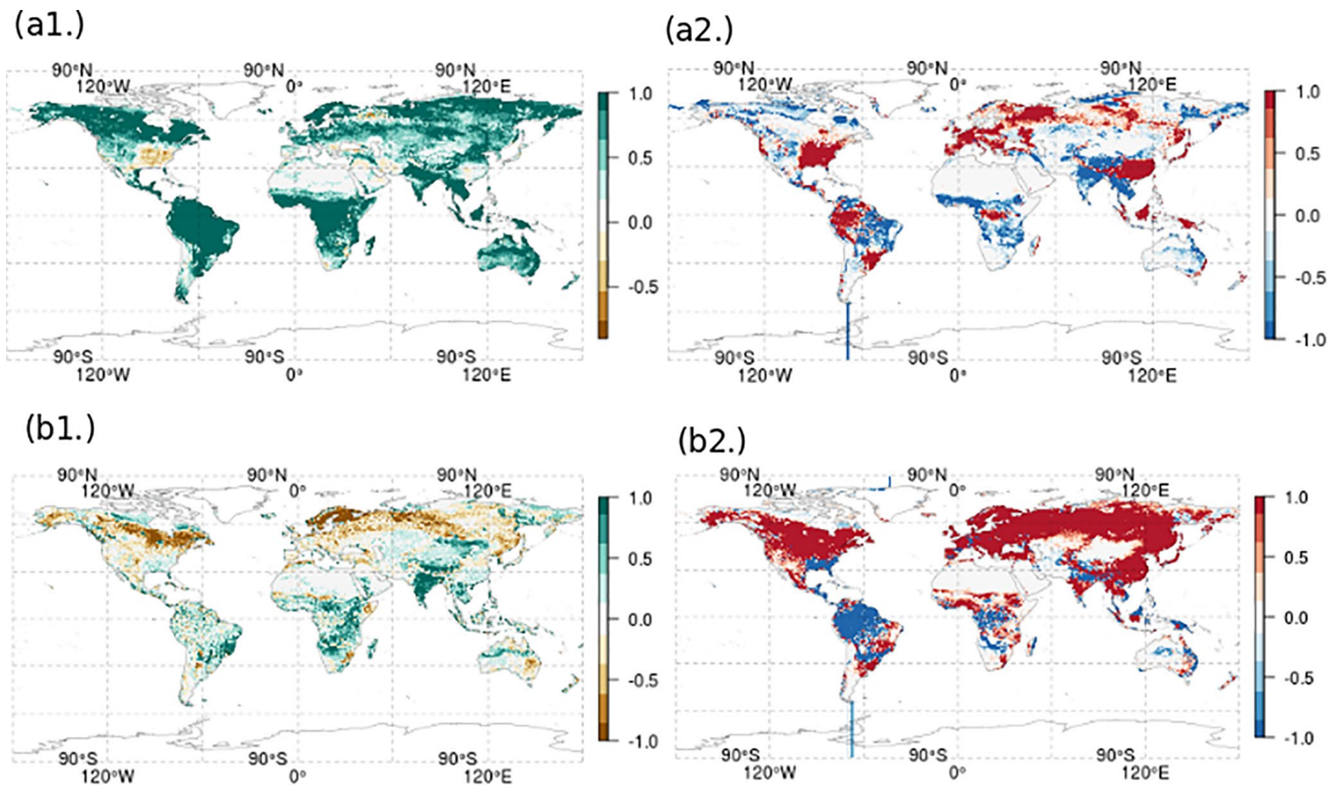

Figure 2. Changes in terrestrial net primary production (NPP) (a1 and b1) and N losses (a2 and b2) in response to RCP 8.5 over the period of 1901–2101, that is, linear trend values of ΔNPP ($gCm^{-2} yr^{-1}$) (left column, i.e., a1 and b1) and ΔNleach (1000 $gNm^{-2} yr^{-1}$) (right column, i.e., a2 and b2) for Background N inputs (rock N + biological N fixation) (top row, i.e., a1 and a2) and N deposition (bottom row, i.e., b2 and b2). For individual N inputs, that is, rock N and biological N fixation separately, see Figure S9.

N and rock weathering (Falkowski et al., 2000). Additional work could focus on background conditions of multiple element interactions, feedback and their influence over the C cycle and climate change.

An important ommission from our model analysis deals with direct interactions among different N sources, particularly between N fixation and N inputs., Studies have shown that N fixation generally increases with experimental N inputs at high latitudes; however, tropical forests maintain a level of high N cycling and "N richness" that doesn't seem to limit N fixation rates (Houlton et al., 2008; Vitousek et al., 2002). While there is a general concensus that increased atmospheric deposition of N reduces biological N fixation (Vitousek et al., 2002), this relationship is not linear and not uniform across ecosystems and types of N fixers (Zheng et al., 2019). Whether rising N deposition would trigger large reductions in N fixation, and thus influence the responsiveness of the terrestrial C cycle to N deposition, is an important question for further work. Moreover, from a preindustrial perspective, both rock N and weathering fluxes have been important over geo-evolutionary time scales, and so future studies attempting to dissect the impacts of the two N inputs will need to carry out simulations in long model spin-up cycles. Although much more work is needed, there is evidence that increased supply of N from bedrock weathering stimulates biological N fixation through feedbacks in the C cycle (Dynarski et al., 2019).

4. Conclusions and Implications

Our study has implications for policies aimed at reducing human N pollution and managing the sustainability of Earth for human prosperity (Hungate et al., 2003; Rockström et al., 2009). Global biogeochemical models have consistently highlighted N deposition's positive influence on NPP, implying that reductions in N deposition will reduce terrestrial CO_2 capture-an unwanted side-effect of reductions in N pollution (Gu et al., 2018). However, results from field experiments reveal both positive and negative NPP responses to N deposition and N fertilizer additions (Thomas et al., 2010). Conversely, we suggest that environmental v

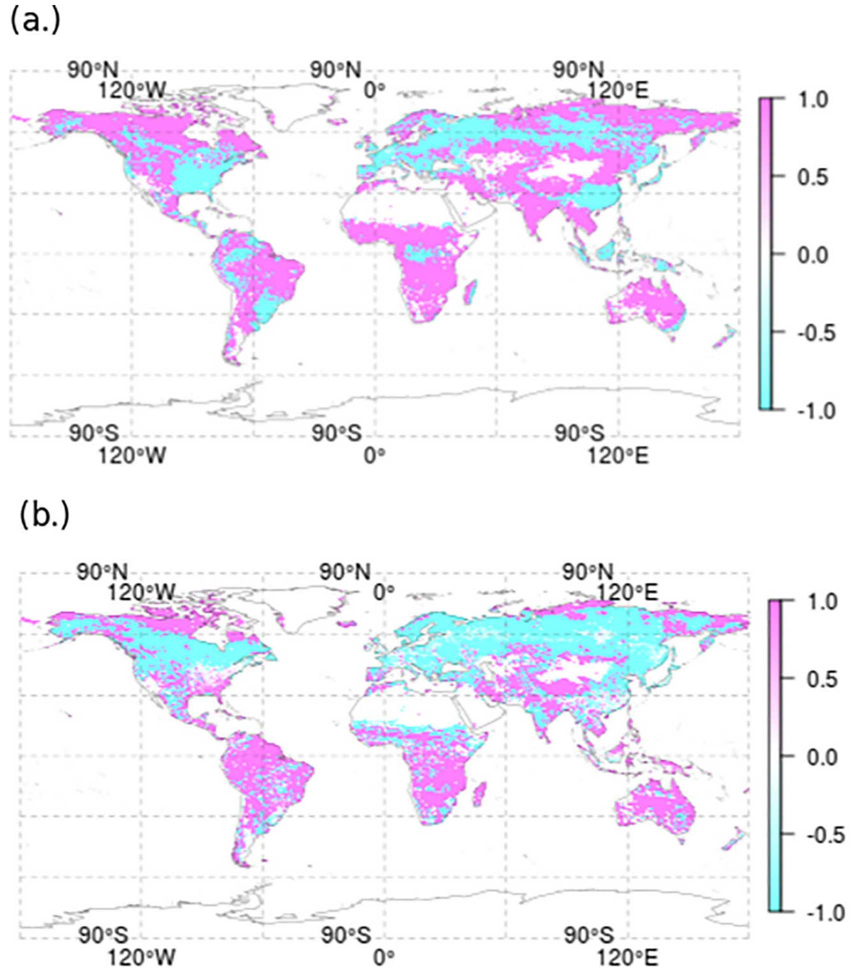

Figure 3. Temporal change in net primary production (ΔNPP):ΔNleach ratio from 1901 to 2101 (linear trend) for (a) background N inputs (rockN + biological N fixation) and (b) atmospheric N deposition. Units are gC/m^2/yr:1000gN/m^2/yr. Positive values (magenta) indicate the degree of utilization of N for NPP while negative values (cyan) imply lack of utilization of N for NPP gains, hence a direct N loss. For individual N inputs, that is, rock N and biological N fixation separately, see Figure S10.

N pollution abatement holds many benefits for the economy, human health and environment (Galloway et al., 2004), without markedly reducing terrestrial C sinks. Given the role of background N input pathways (rock N weathering and biological N fixation) in driving historical and future C gains, we suggest that land conservation priorities might consider focus on preserving C sinks where these N inputs are naturally high, such as biologically diverse tropical forests, mountainous regions, and the rapidly changing boreal zone.

Data Availability Statement

We used climate data from Climatic Research Unit (CRU) (Jones & Harris, 2013) which is based on observations of spatial resolution at 0.5° latitude and longitude. Downloaded from https://catalogue.ceda.ac.uk/uuid/89e1e34ec3554dc98594a5732622bce9. For future projections, we used climate trends of the GCM (general circulation model) FGOALS-g2 (Li et al., 2013). Downloaded from https://cera-www.dkrz.de/WDCC/ui/cerasearch/q. Atmospheric CO_2 concentrations were downloaded from: http://www.pik-potsdam.de/~mmalte/rcps/ and https://www.iiasa.ac.at/web-apps/tnt/RcpDb/dsd?Action=htmlpage&page=download. With regards to the N inputs, Monthly atmospheric N-deposition rates were taken from the ACCMIP database (Lamarque et al., 2013). Downloaded from http://acd.ucar.edu/~lamar/ACCMIP/Deposition/all_fields_062013.tar.gz. Biological N fixation is computed using the linear regression equation for predicting N fixation values (Cleveland et al., 1999) as based on actual evapotranspiration data (AET). Observed AET data were taken from MODIS-MOD16 (Mu et al., 2007) future trends were taken from GCM FGOALS-g2 (Li et al., 2013) downloaded from http://files.ntsg.umt.edu/data/NTSG_Products/MOD16/. N flux from bedrock weathering (B. Z. Houlton et al., 2018) has been shared by the authors in a public data repository as Dass et al. (2021) and can be downloaded using the link: https://datadryad.org/stash/dataset/doi:10.5061/dryad.5x69p8d1x. The LPJ-GUESS(Smith et al., 2014) version 3.1, a dynamic global vegetation model (DGVM) that we used is available from http://web.nateko.lu.se/lpj-guess/. The Net Primary Productivity and N leaching flux model outputs (spatial) for the pre-industrial period as well as for the 20th and 21st centuries have been shared by the authors in a public data repository as Dass et al. (2021) and can be downloaded using the link: https://datadryad.org/stash/dataset/doi:10.5061/dryad.5x69p8d1x.

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