

**Estimating the CO<sub>2</sub> fertilization effect on extratropical forest productivity from Flux-tower observations**

Chunhui Zhan<sup>1,2</sup>, René Orth<sup>1,3</sup>, Hui Yang<sup>1</sup>, Markus Reichstein<sup>1</sup>, Sönke Zaehle<sup>1</sup>, Martin G. De Kauwe<sup>4</sup>, Anja Rammig<sup>2</sup> and Alexander J. Winkler<sup>1</sup>

<sup>1</sup>Max Planck Institute for Biogeochemistry, 07745 Jena, Germany, <sup>2</sup>School of Life Sciences, Technical University of Munich, 85354 Freising, Germany, <sup>3</sup>Faculty of Environment and Natural Resources, University of Freiburg, 79106 Freiburg, Germany, <sup>4</sup>School of Biological Sciences, University of Bristol, BS8 1TQ Bristol, UK

**Contents of this file**

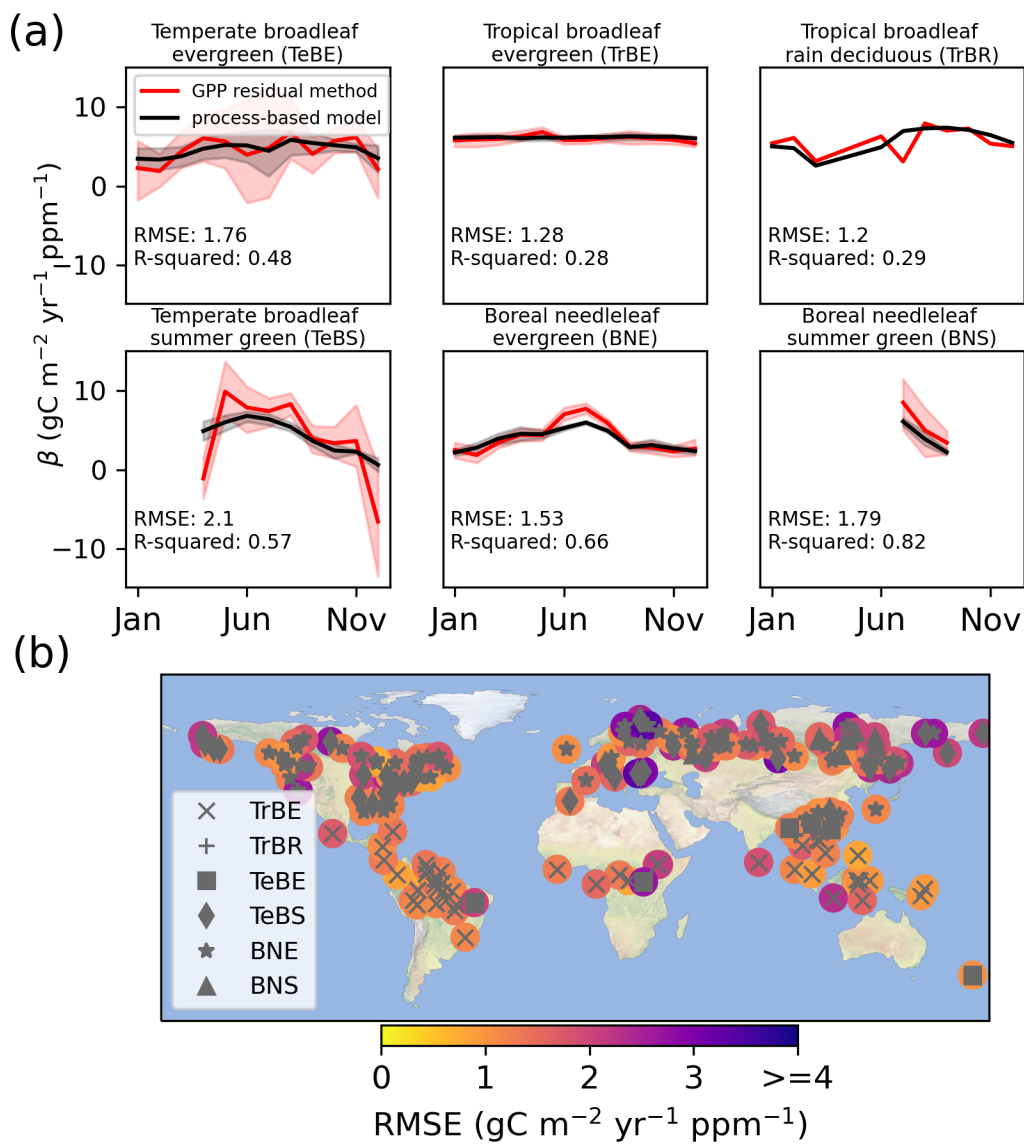
Figures S1 to S14

Tables S1 to S5

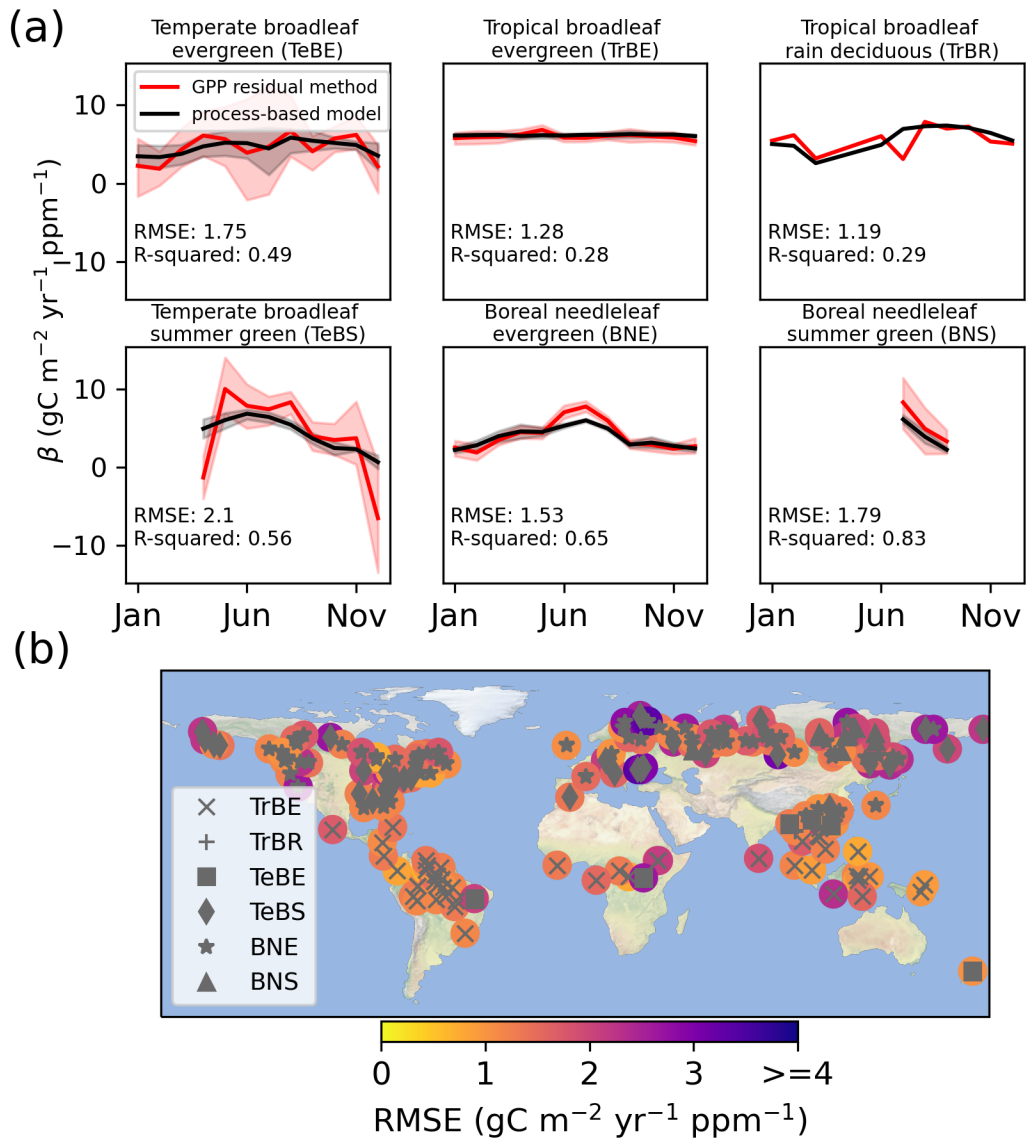
Supplementary text S1

**Introduction**

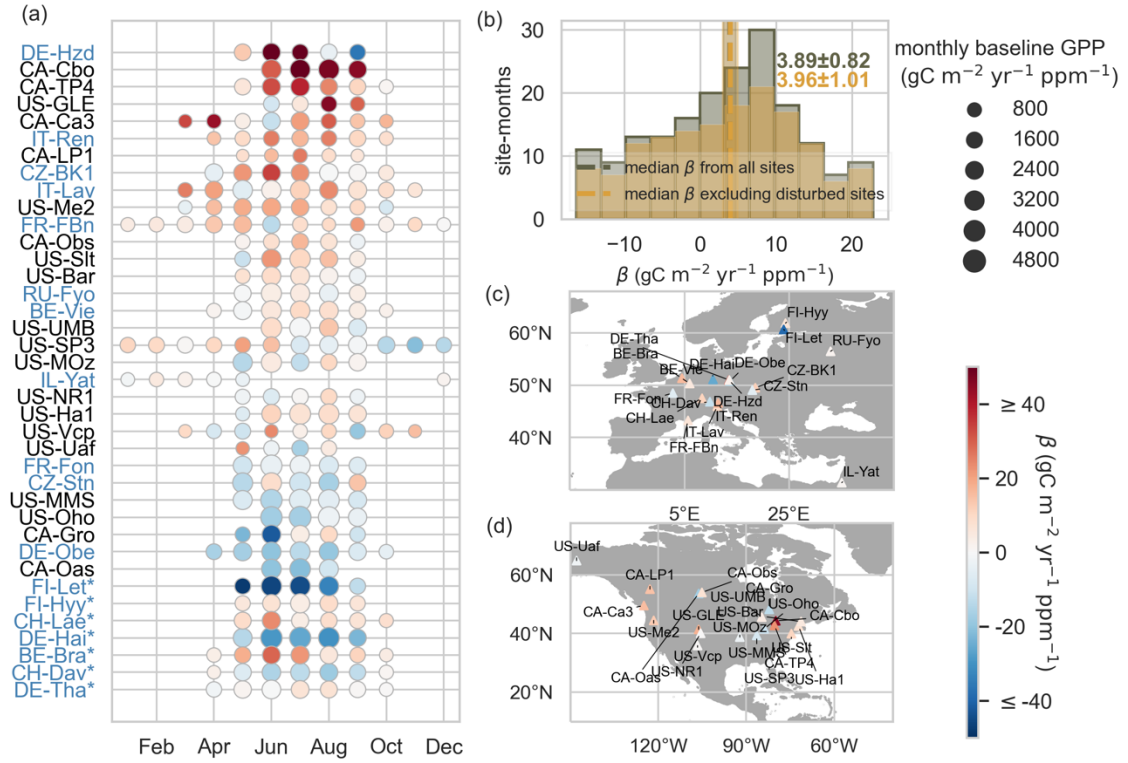
This supporting information provides 1) the estimation of  $\beta$  and  $\gamma$  effects using different statistical models; 2) tables for information about eddy covariance sites and TRENDY simulations; 3) tests of the statistical method to incorporate the cumulative effect of nitrogen deposition ( $N_{\text{dep}}$ ) to the  $\beta$  estimation.



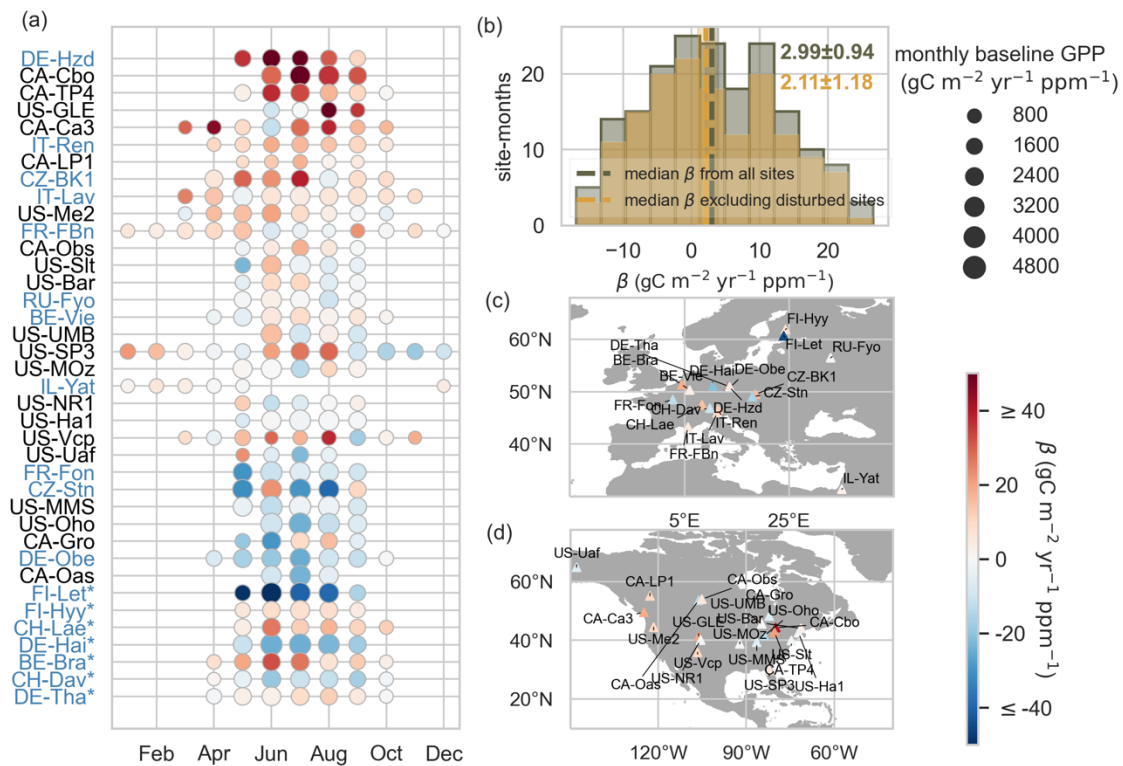
**Figure S1.** Validation of the statistical method with QUINCY simulations. Analogous to Figure 2 but  $\beta$  is estimated by multivariate regression model.



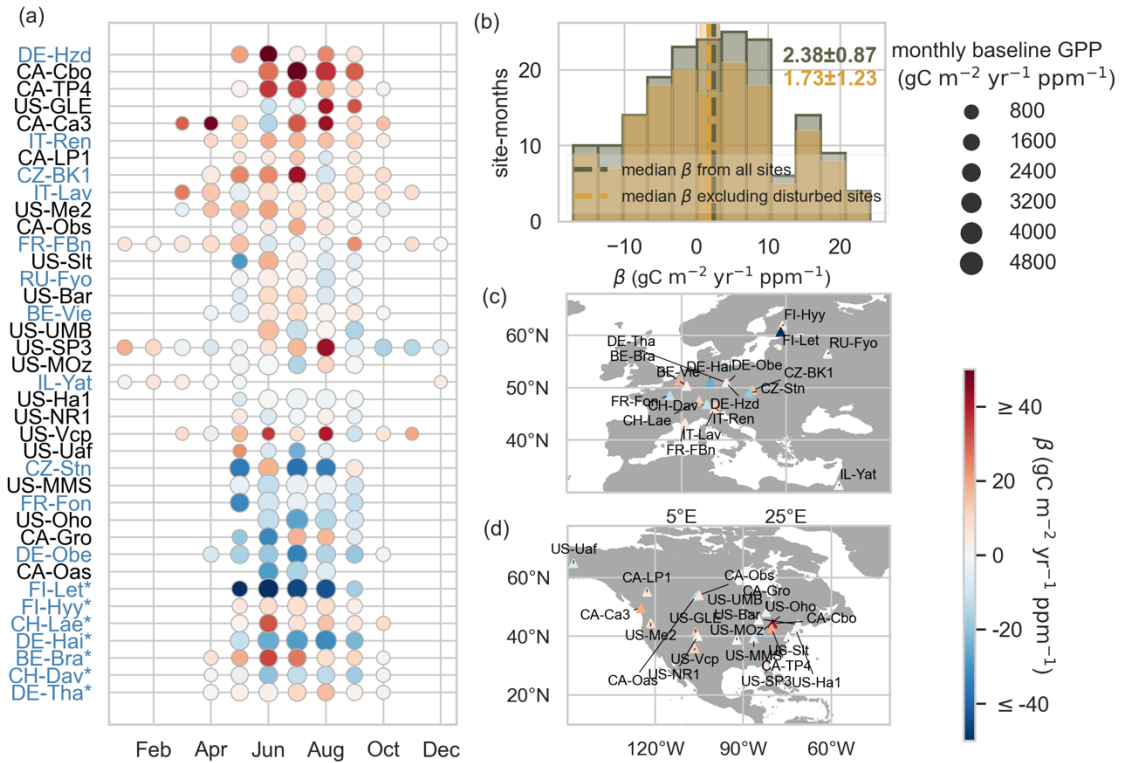
**Figure S2.** Validation of the statistical method with QUINCY simulations. Analogous to Figure 2 but  $\beta$  is estimated by a simple multivariate regression model.



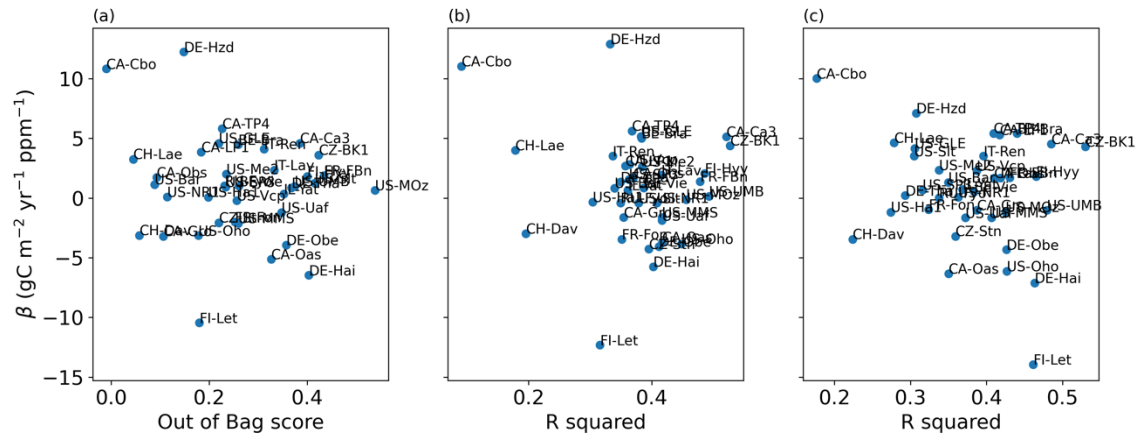
**Figure S3.** Estimation of  $\beta$  from eddy covariance dataset. Analogous to Figure 3 but nitrogen deposition data from the forcing database for CMIP6 models is included as a predictor of GPP (Methods). Sites in the panel (a) are shown the same order as Figure 3.



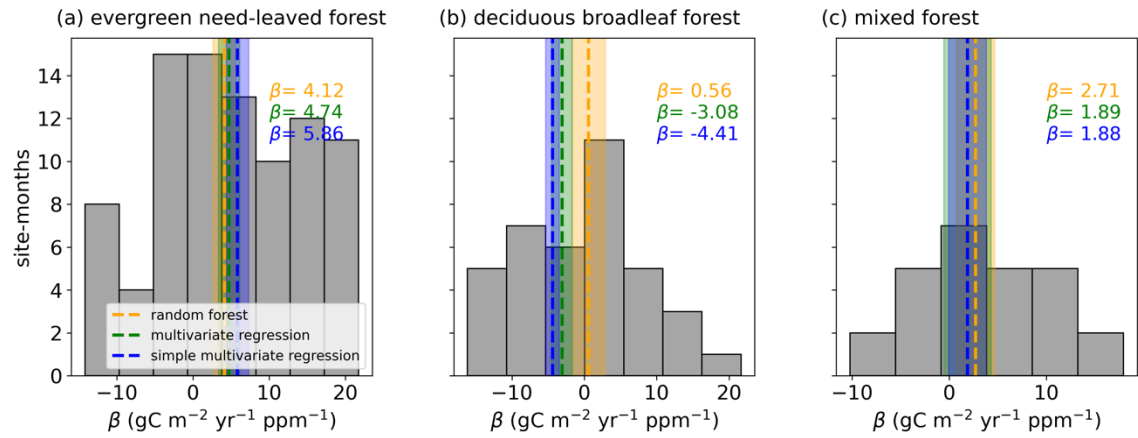
**Figure S4.** Estimation of  $\beta$  from eddy covariance dataset. Analogous to Figure 3 but  $\beta$  is estimated by a multivariate linear regression model. Sites in the panel (a) are shown the same order as Figure 3.



**Figure S5.** Estimation of  $\beta$  from eddy covariance dataset. Analogous to Figure 3 but  $\beta$  is estimated by the simple multivariate linear regression model (Methods). Sites in the panel (a) are shown the same order as Figure 3.

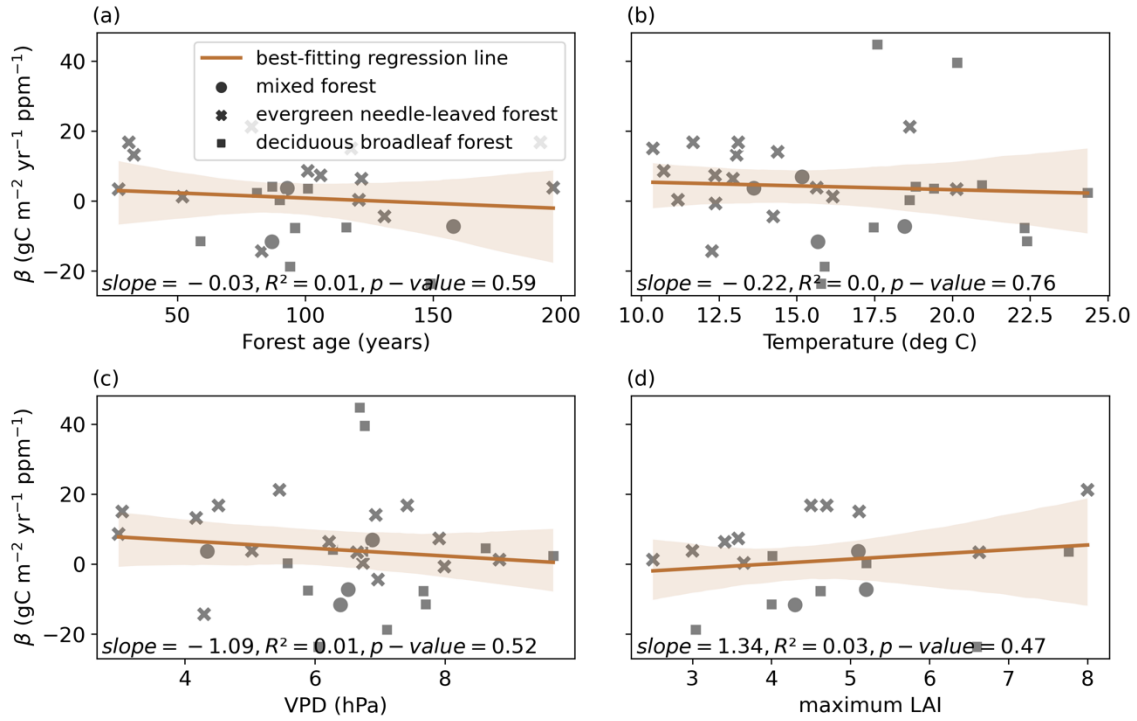


**Figure S6.** The performance of (a) the random forest model (Out of Bag; OOB score), (b) the multivariate regression model (R squared) and (c) the simple multivariate regression model (R squared) against estimated annual  $\beta$  at each site.

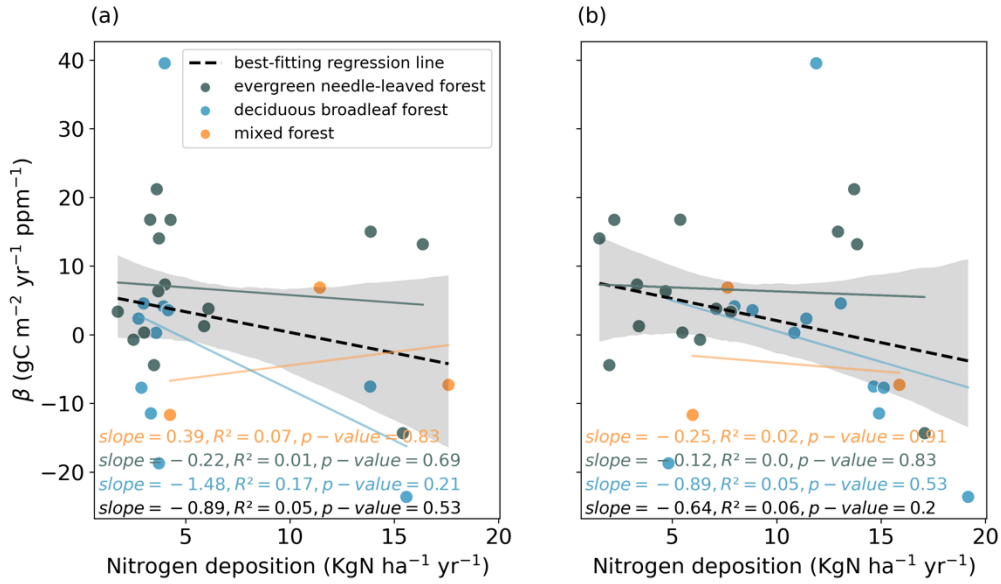


**Figure S7.** Histogram plots of  $\beta$  estimated for (a) evergreen needleleaf forest (b) deciduous broadleaf forest and (c) mixed forest in eddy covariance dataset. The vertical dashed lines denote the median  $\beta$  estimated from three methods (random forest model in yellow, multivariate regression model in green, simple multivariate regression model in blue). The shaded area indicates the bootstrap estimates for the uncertainty of  $\beta$  correspondingly.

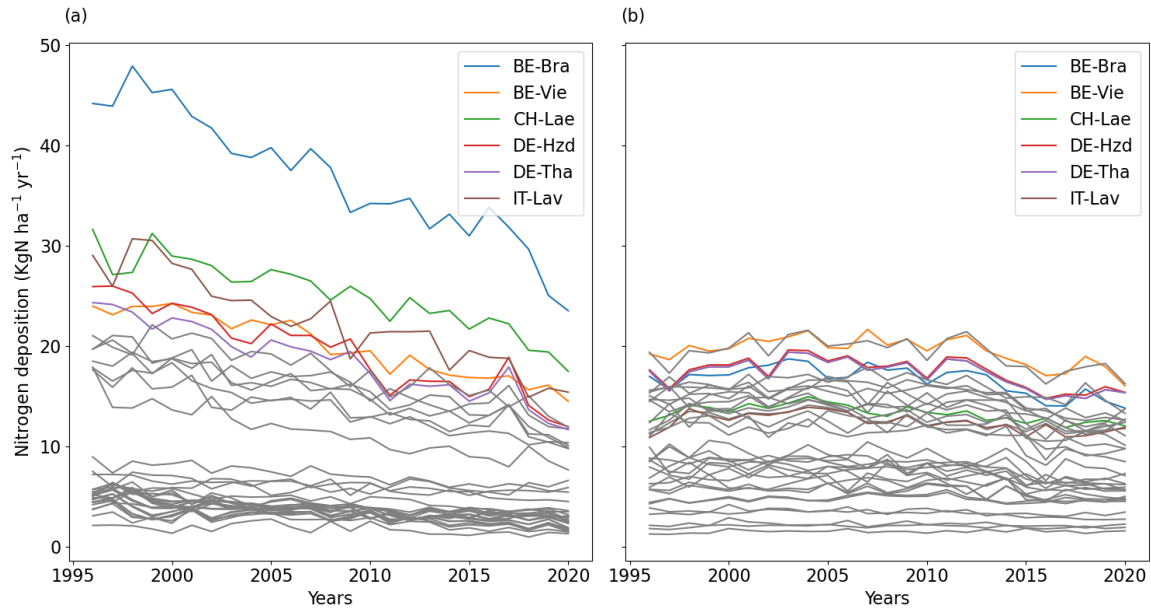




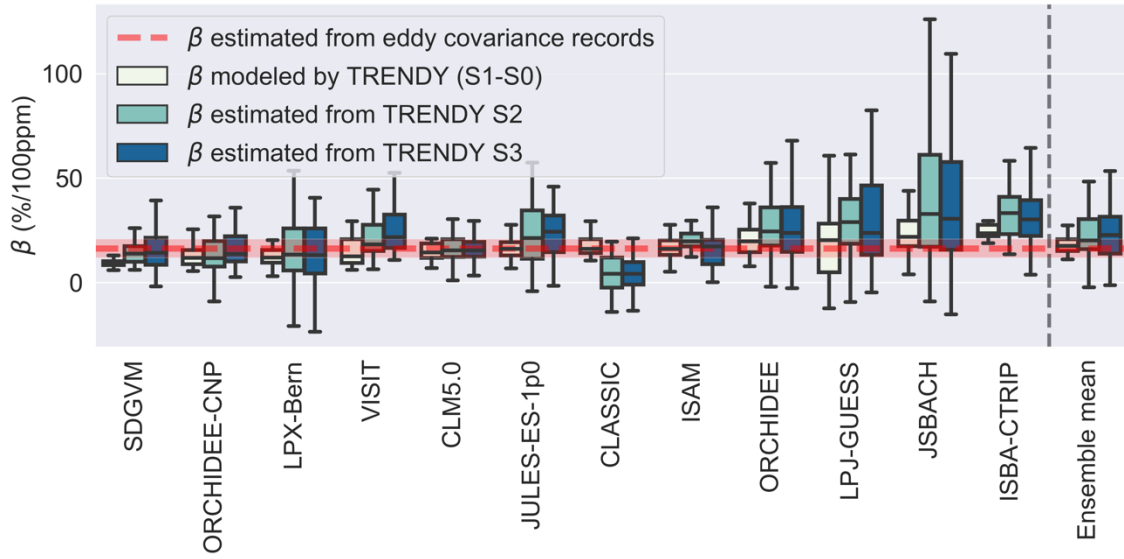
**Figure S8.** Scatter plot (with best-fitting regression line) and correlation between  $\beta$  and (a) forest age, (b) mean temperature in growing seasons, (c) mean vapor pressure deficit in growing seasons and (d) maximum leaf area index. The shaded area represents the 95% confidence interval around the regression line. Circles, squares and cross represent mixed forest, evergreen needle-leaved forest, deciduous broadleaf forest.



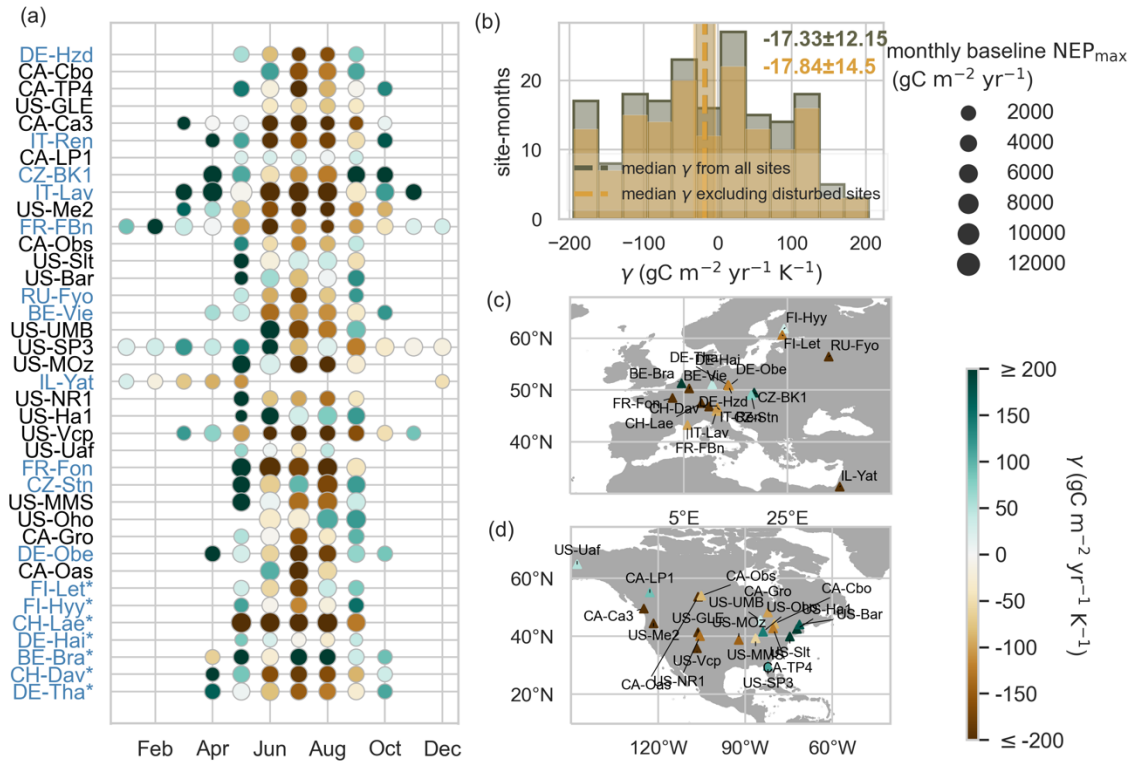
**Figure S9.** Scatter plot (with best-fitting regression line) and correlation between  $\beta$  and nitrogen deposition data from (a) the European Monitoring and Evaluation Programme (EMEP) Meteorological Synthesizing Centre - West (MSC-W) model, and (b) the forcing database for CMIP6 models. The orange, green, and blue circles represent mixed forest, evergreen needle-leaved forest, and deciduous broadleaf forest, respectively. The best-fitting regression lines for different forest types are depicted in corresponding colors. The shaded area represents the 95% confidence interval around the regression line fitted across all sites.



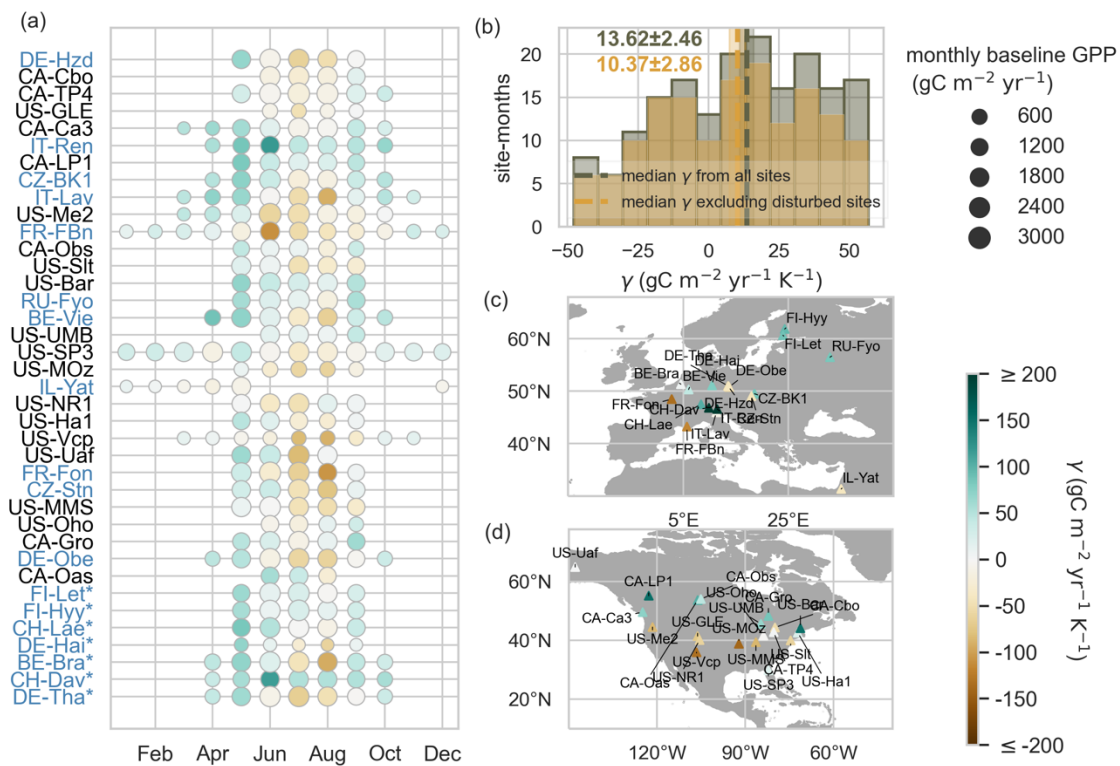
**Figure S10.** Temporal evolution of Nitrogen deposition ( $N_{\text{dep}}$ ) near the 38 forest sites from 1996 to 2020, using data from (a) the European Monitoring and Evaluation Programme (EMEP) Meteorological Synthesizing Centre - West (MSC-W) model, and (b) the forcing database for CMIP6 models. In instances where the annual  $N_{\text{dep}}$  remains below  $24 \text{ KgN ha}^{-1} \text{ yr}^{-1}$  (Methods) throughout the entire period in panel (a), the lines are depicted in grey. Conversely, when the  $N_{\text{dep}}$  exceeds this threshold, the lines are presented in different colors. Specifically,  $N_{\text{dep}}$  data near the sites BE-Bra, BE-Vie, CH-Lai, DE-Hzd, DE-Tha, and IT-Lav are distinguished by unique colors. Although the annual  $N_{\text{dep}}$  values in panel (b) consistently fall below  $24 \text{ KgN ha}^{-1} \text{ yr}^{-1}$ , the color scheme is maintained in panel (a) for consistency.



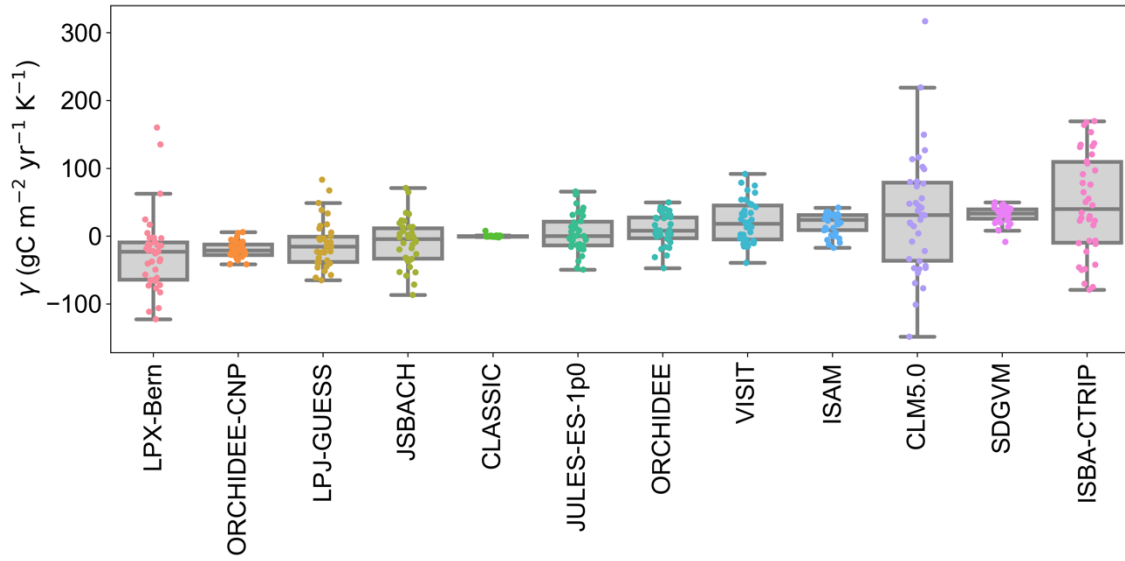
**Figure S11.** Comparing  $\beta$  estimated from eddy covariance data and the TRENDY model ensemble. Analogous to Figure 5 but  $\beta$  is estimated as the relative change in GPP per 100 ppm increase in atmospheric CO<sub>2</sub> concentration (Methods).



**Figure S12.** Estimation of  $\gamma$  from eddy covariance dataset using the *GPP residual method* with a random forest model. Analogous to Figure 6 but  $\gamma$  is estimated for  $\text{NEP}_{\text{max}}$ .



**Figure S13.** Estimation of  $\gamma$  from TRENDY ensemble mean. Analogous to Figure 6 but  $\gamma$  is estimated from the TRENDY ensemble.



**Figure S14.** Estimation of  $\gamma$  from the TRENDY ensemble. The medians and interquartile ranges of  $\gamma$  are shown for each model, as horizontal lines within boxes, the upper and bottom lines of the box, respectively. Box plots for individual models are ordered according to the median  $\gamma$  across models. The points represent the mean  $\gamma$  at grid-cells containing the 38 considered eddy covariance sites for each model.

**Table S1.** List of eddy covariance sites used in this study. Plant functional type (IGBP class), coordinates, and data citation are reported.

Site code	PFT	Latitude	Longitude	Data citation
BE-Bra	MF	51.3092	4.5206	<a href="https://doi.org/10.18160/YVQB-K6WF">https://doi.org/10.18160/YVQB-K6WF</a>
BE-Vie	MF	50.3051	5.9981	<a href="https://doi.org/10.18160/DF9X-QMRK">https://doi.org/10.18160/DF9X-QMRK</a>
CA-Ca3	ENF	49.5346	-124.9004	<a href="https://doi.org/10.17190/AMF/1480302">https://doi.org/10.17190/AMF/1480302</a>
CA-Cbo	DBF	44.3167	-79.9333	<a href="https://doi.org/10.17190/AMF/1854365">https://doi.org/10.17190/AMF/1854365</a>
CA-Gro	MF	48.2167	-82.1556	<a href="https://doi.org/10.17190/AMF/1902823">https://doi.org/10.17190/AMF/1902823</a>
CA-LP1	ENF	55.1119	-122.8414	<a href="https://doi.org/10.17190/AMF/1832155">https://doi.org/10.17190/AMF/1832155</a>
CA-TP4	ENF	42.7102	-80.3574	<a href="https://doi.org/10.17190/AMF/1246012">https://doi.org/10.17190/AMF/1246012</a>
CA-Oas	DBF	53.6289	-106.1978	<a href="https://doi.org/10.17190/AMF/1375197">https://doi.org/10.17190/AMF/1375197</a>
CA-Obs	ENF	53.9872	-105.1178	<a href="https://doi.org/10.17190/AMF/1375198">https://doi.org/10.17190/AMF/1375198</a>
CH-Dav	ENF	46.8153	9.8559	<a href="https://doi.org/10.18160/1JA9-VJEV">https://doi.org/10.18160/1JA9-VJEV</a>
CH-Lae	MF	47.4781	8.3650	<a href="https://doi.org/10.18160/51Z6-S5XF">https://doi.org/10.18160/51Z6-S5XF</a>
CZ-BK1	ENF	49.5021	18.5369	<a href="https://doi.org/10.18160/2X51-1SD0">https://doi.org/10.18160/2X51-1SD0</a>
CZ-Stn	DBF	49.0360	17.9699	<a href="https://doi.org/10.18160/0JY0-ZCD6">https://doi.org/10.18160/0JY0-ZCD6</a>
DE-Hai	DBF	51.0792	10.4522	<a href="https://doi.org/10.18160/CR66-PJ24">https://doi.org/10.18160/CR66-PJ24</a>
DE-Hzd	DBF	50.9638	13.4898	<a href="https://doi.org/10.18160/2G60-ZHAK">https://doi.org/10.18160/2G60-ZHAK</a>
DE-Obe	ENF	50.7836	13.7196	<a href="https://doi.org/10.18140/FLX/1440151">https://doi.org/10.18140/FLX/1440151</a>
DE-Tha	ENF	50.9636	13.5669	<a href="https://doi.org/10.18160/8FBV-1K18">https://doi.org/10.18160/8FBV-1K18</a>
FI-Hyy	ENF	61.8475	24.2950	<a href="https://doi.org/10.18160/XTBV-XCJV">https://doi.org/10.18160/XTBV-XCJV</a>
FI-Let	ENF	60.6418	23.9595	<a href="https://doi.org/10.18140/FLX/1440227">https://doi.org/10.18140/FLX/1440227</a>
FR-FBn	MF	43.2408	5.6787	<a href="https://doi.org/10.18160/KGN6-K1CX">https://doi.org/10.18160/KGN6-K1CX</a>
FR-Fon	MF	48.4764	2.7801	<a href="https://doi.org/10.18160/X1J0-H684">https://doi.org/10.18160/X1J0-H684</a>
IL-Yat	ENF	31.3450	35.0520	<a href="https://doi.org/10.18160/MAGT-CWRW">https://doi.org/10.18160/MAGT-CWRW</a>
IT-Lav	ENF	45.9553	11.2812	<a href="https://doi.org/10.18160/HZSQ-G19C">https://doi.org/10.18160/HZSQ-G19C</a>
IT-Ren	ENF	46.5869	11.4337	<a href="https://doi.org/10.18160/WMCA-8P4P">https://doi.org/10.18160/WMCA-8P4P</a>
RU-Fyo	ENF	56.4617	32.9239	<a href="https://doi.org/10.18160/XMER-D4NR">https://doi.org/10.18160/XMER-D4NR</a>
US-Bar	DBF	44.0646	-71.2881	<a href="https://doi.org/10.17190/AMF/2006969">https://doi.org/10.17190/AMF/2006969</a>
US-GLE	ENF	41.3665	-106.2399	<a href="https://doi.org/10.17190/AMF/1871136">https://doi.org/10.17190/AMF/1871136</a>



US-Ha1	DBF	42.5378	-72.1715	<a href="https://doi.org/10.17190/AMF/1871137">https://doi.org/10.17190/AMF/1871137</a>
US-Me2	ENF	44.4523	-121.5574	<a href="https://doi.org/10.17190/AMF/1854368">https://doi.org/10.17190/AMF/1854368</a>
US-MMS	DBF	39.3232	-86.4131	<a href="https://doi.org/10.17190/AMF/1854369">https://doi.org/10.17190/AMF/1854369</a>
US-MOz	DBF	38.7441	-92.2000	<a href="https://doi.org/10.17190/AMF/1854370">https://doi.org/10.17190/AMF/1854370</a>
US-NR1	ENF	40.0329	-105.5464	<a href="https://doi.org/10.17190/AMF/1871141">https://doi.org/10.17190/AMF/1871141</a>
US-Oho	DBF	41.5545	-83.8438	<a href="https://doi.org/10.17190/AMF/1246089">https://doi.org/10.17190/AMF/1246089</a>
US-Slt	DBF	39.9138	-74.5960	<a href="https://doi.org/10.17190/AMF/1246096">https://doi.org/10.17190/AMF/1246096</a>
US-SP3	ENF	29.7548	-82.1633	<a href="https://doi.org/10.17190/AMF/1246102">https://doi.org/10.17190/AMF/1246102</a>
US-Uaf	ENF	64.8663	-147.8555	<a href="https://doi.org/10.17190/AMF/1480322">https://doi.org/10.17190/AMF/1480322</a>
US-UMB	DBF	45.5598	-84.7138	<a href="https://doi.org/10.17190/AMF/2204882">https://doi.org/10.17190/AMF/2204882</a>
US-Vcp	ENF	35.8624	-106.5974	<a href="https://doi.org/10.17190/AMF/1246122">https://doi.org/10.17190/AMF/1246122</a>

---

**Table S2.** Variables from eddy covariance dataset used in this study.

<b>Variables</b>	<b>Description</b>	<b>Time scale</b>	<b>Unit</b>
NEE_VUT_USTAR50	Net Ecosystem Exchange, using Variable Ustar Threshold (VUT) for each year, from 50 percentile of USTAR threshold, half-hourly data	Half-hourly	$\mu\text{mol m}^{-2} \text{s}^{-1}$
GPP_NT_VUT_USTAR50	Gross Primary Production, from Nighttime partitioning method, based on NEE_VUT_USTAR50, calculated from half-hourly data	daily	$\text{gC m}^{-2} \text{day}^{-1}$
TA_F_DAY	Averaged daytime air temperature	daily	deg C
VPD_F_MDS	Vapor Pressure Deficit, gapfilled using MDS method, average from half-hourly data	daily	hPa
P_F	Sum of precipitation from half-hourly data	daily	$\text{mm day}^{-1}$
LE_F_MDS	Latent heat flux, gapfilled using MDS method, average from half-hourly data	daily	$\text{W m}^{-2}$
SW_IN_F_MDS	Shortwave radiation, incoming, gapfilled using MDS (negative values set to zero, e.g., negative values from instrumentation noise), average from half-hourly data	daily	$\text{W m}^{-2}$

**Table S3.** Simulations from TRENDY v9 used in this study.

<b>Simulations</b>	<b>CO<sub>2</sub> concentration</b>	<b>Climate</b>	<b>LULCCs forcing</b>
S0	Pre-industrial	Pre-industrial	Pre-industrial
S1	Observed	Pre-industrial	Pre-industrial
S2	Observed	Observed	Pre-industrial
S3	Observed	Observed	LUH2/HYDE

## Supplementary text S1. Testing the statistical method to incorporate the cumulative effect of nitrogen deposition ( $N_{dep}$ ) to the $\beta$ estimation

We develop the so-called *GPP residual method* that statistically captures the sensitivity of GPP to  $CO_2$  and climate variables at different time scales to account for co-linearities among the drivers (Methods). We note that the GPP residual may also include other long-term effects specific to individual sites, such as signals related to nitrogen deposition ( $N_{dep}$ ). Similar to the increasing  $CO_2$  effect, the response to  $N_{dep}$  change is likely a long-term phenomenon and might even show some degree of hysteresis, associated with the slow-turnover pools of nitrogen (N) in wood and soil (Gilliam et al., 2019). The input of  $N_{dep}$  could be taken up by vegetation and microbes and subsequently redistributed within the plant-soil system, or be retained or stored in the soil or biomass for extended periods before being released or lost through various processes (Galloway et al., 2004). Thus, it's crucial to account for the cumulative effect of  $N_{dep}$  that undergoes cycling within the natural system. Assuming a long residence time of reactive nitrogen in the system (Gruber & Galloway, 2008), the accumulated  $N_{dep}$  at year  $t$  in this analysis can be considered as:

$$Accumulated\ N_{dep_t} = \sum_{i=1}^t N_{dep_i} \quad (1)$$

Where  $N_{dep_i}$  is the mean  $N_{dep}$  at year  $i$ . We also explored applying a low-pass filter to the input data when calculating accumulated  $N_{dep}$  at a later stage (Equation 2, 3), to capture the dynamic of N cycling. Our statistical method aims to separate the short-term responses (e.g., temperature, VPD, etc.) from long-term responses (e.g.,  $CO_2$ ). There may be an opportunity to take one more step to further isolate the effects of  $CO_2$  and accumulated  $N_{dep}$  from these long-term responses. However, this is challenging as  $CO_2$  and accumulated  $N_{dep}$  may be correlated at a long-term scale, which complicates the task of separating these two factors using the linear regression models. We assess the feasibility of integrating accumulated  $N_{dep}$  into our *GPP residual method* using both synthetic data and eddy covariance record.

We conduct three sets of tests (Table S4) to integrate the cumulative effect of  $N_{dep}$  ( $\eta$ ) into the linear regression model of  $GPP_{residual}$  after eliminating the climate effect using the GPP residual method (Figure 1d and Methods). Within each set of tests, we employ five linear regression models to examine the influence of co-linearity among independent variables and to determine if the cumulative effect of  $N_{dep}$  can be effectively isolated. Specifically, the five linear regression models are designed as: (1) M1:  $GPP_{residual} \sim CO_2$ . The linear model M1 is used to calculate the  $CO_2$  fertilization effect ( $\beta$ ) as the slope in the main text; (2) M2:  $GPP_{residual} \sim CO_2 + N_{dep}$  calculates both  $\beta$  and  $\eta$  that are coefficients associated with  $CO_2$  and accumulated  $N_{dep}$  (referred to as  $N_{dep}$  in the linear models for simplicity), respectively; (3) M3:  $GPP_{residual} \sim CO_2 + years$  is employed to calculate the coefficient  $\beta$  amidst a linear sequence (here years) within the linear regression model. This is an examination of whether the linear regression model accurately attributes effects when co-linearity among independent variables is anticipated; (4) M4:  $GPP_{residual} \sim N_{dep}$  attributes all long-term effects on GPP to accumulated  $N_{dep}$  as  $\eta$ . We assess whether accumulated  $N_{dep}$ , as the sole independent variable, explains the majority of the variance in  $GPP_{residual}$ ; (5) M5:  $GPP_{residual} \sim years$  calculates the trend of  $GPP_{residual}$  as the slope. The accumulated  $N_{dep}$  (Equation 1) is used in M2 and M4 linear regression models in all three tests.

The distinctions among the three sets of tests lie in the derivation of the  $GPP_{residual}$ . In **Test #1**, the dependent variable  $GPP_{residual}$  is modeled by the terrestrial biosphere model QUINCY, which is used in the main text to develop and evaluate the GPP residual method (Methods).  $GPP_{residual}$  is calculated as the difference between simulations forced with transient  $CO_2$  and constant  $CO_2$ , reflecting the theoretical comprehension of the  $CO_2$  fertilization effect. Consequently, within the

test set,  $\beta$  in M1, M2 and M3 should be comparable if the linear regression models can successfully isolate the CO<sub>2</sub> effect, even with different combinations of independent variables. The R<sup>2</sup> in M4 is expected to be low since no N<sub>dep</sub> effect is encoded in the QUINCY GPP<sub>residual</sub>.

In **Test #2**, GPP<sub>residual</sub> is derived from the QUINCY simulation forced with transient CO<sub>2</sub> using the GPP residual method (Methods). Given that the GPP residual method can only separate the short-term climate effect with other long-term effects, the GPP<sub>residual</sub> may encompass other long-term effects (such as long-term GPP acclimation to climate) besides the CO<sub>2</sub> effect. These effects are likely to be attributed to CO<sub>2</sub> or other drivers exhibiting a trend by the linear regression model. However, since nitrogen dynamics are not accounted in these simulations, the R<sup>2</sup> in M4 is anticipated to be low.

In **Test #3**, GPP<sub>residual</sub> is derived from the eddy covariance (EC) dataset using the GPP residual method. Potentially, there could be other long-term effects present in GPP<sub>residual</sub> aside from CO<sub>2</sub>, such as the impact of N<sub>dep</sub>.

**Test #1** and **Test #2** involve 166 synthetic sites in the QUINCY experiments, serving as the proof-of-concept as discussed in the main text. **Test #3** incorporates 38 EC sites used for estimating  $\beta$  and  $\gamma$ , also discussed in the main text.

**Table S4.** Linear regression models of the GPP<sub>residual</sub> used to test the cumulative effect of N<sub>dep</sub> calculated by equation 1

<b>Test #1 GPP<sub>residual</sub> modeled by QUINCY</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> (gC m <sup>-2</sup> yr <sup>-1</sup> ppm <sup>-1</sup> )	<b>Median <math>\eta</math></b> (KgC KgN <sup>-1</sup> )	<b>Median trend</b> (gC m <sup>-2</sup> yr <sup>-2</sup> )	<b>Mean R<sup>2</sup></b>
<b>M1:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub>	5.78	-	-	0.718
<b>M2:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + N <sub>dep</sub>	4.32	4.55	-	0.727
<b>M3:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + years	3.95	-	3.37	0.728
<b>M4:</b> GPP <sub>residual</sub> ~ N <sub>dep</sub>	-	25.88	-	0.715
<b>M5:</b> GPP <sub>residual</sub> ~ years	-	-	12.14	0.717
<b>Test #2 GPP<sub>residual</sub> derived by the GPP residual method in QUINCY simulation</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> (gC m <sup>-2</sup> yr <sup>-1</sup> ppm <sup>-1</sup> )	<b>Median <math>\eta</math></b> (KgC KgN <sup>-1</sup> )	<b>Median trend</b> (gC m <sup>-2</sup> yr <sup>-2</sup> )	<b>Mean R<sup>2</sup></b>
<b>M1:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub>	5.82	-	-	0.356
<b>M2:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + N <sub>dep</sub>	3.01	10.40	-	0.368
<b>M3:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + years	2.84	-	6.67	0.369
<b>M4:</b> GPP <sub>residual</sub> ~ N <sub>dep</sub>	-	26.77	-	0.356
<b>M5:</b> GPP <sub>residual</sub> ~ years	-	-	12.28	0.356
<b>Test #3 GPP<sub>residual</sub> derived by the GPP residual method in EC dataset</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> (gC m <sup>-2</sup> yr <sup>-1</sup> ppm <sup>-1</sup> )	<b>Median <math>\eta</math></b> (KgC KgN <sup>-1</sup> )	<b>Median trend</b> (gC m <sup>-2</sup> yr <sup>-2</sup> )	<b>Mean R<sup>2</sup></b>
<b>M1:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub>	3.18	-	-	0.100
<b>M2:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + N <sub>dep</sub>	-3.33	20.30	-	0.168
<b>M3:</b> GPP <sub>residual</sub> ~ CO <sub>2</sub> + years	-3.69	-	24.23	0.168
<b>M4:</b> GPP <sub>residual</sub> ~ N <sub>dep</sub>	-	11.77	-	0.145
<b>M5:</b> GPP <sub>residual</sub> ~ years	-	-	13.85	0.144

We find that the mean  $R^2$  in M1, M4 and M5 are comparable in all three sets of tests (Table S4). That means all predictors, here  $\text{CO}_2$ , accumulated  $N_{\text{dep}}$  and years explain an equal proportion of the variance in the  $\text{GPP}_{\text{residual}}$ . This implies that either all predictors are equally important, or the collinearity between predictors create redundancy in the model. As we priorly know that the  $\text{GPP}_{\text{residual}}$  in **Test #1** is modeled by QUINCY simulations and only include the  $\text{CO}_2$  effect, the latter implication is valid. Or in another word, this suggests that the linear regression is likely to misattribute the cumulative  $N_{\text{dep}}$  effect even when there is no actual influence of  $N_{\text{dep}}$  on the dependent variable, simply because  $\text{CO}_2$  and accumulated  $N_{\text{dep}}$  exhibit strong correlation, as any other linear sequence (e.g., years). Statistics in **Test #2** show consistency with **Test #1**, where the misattribution to  $N_{\text{dep}}$  exist when there is no effect of  $N_{\text{dep}}$  in the  $\text{GPP}_{\text{residual}}$ . These findings challenge the linear regression model's capability to effectively handle co-linearity issues. In **Test #3**, the estimation of  $\beta$  by M2 become negative after incorporating accumulated  $N_{\text{dep}}$  into the linear regression model. While it's possible that the  $N_{\text{dep}}$  effect influences the relevance of GPP in **Test #3**, the accuracy of the  $\beta$  estimation is likely compromised due to collinearity issues identified in the previous two tests with QUINCY simulations.

As  $N_{\text{dep}}$  might not have an immediate effect, we adjusted the method of accumulating  $N_{\text{dep}}$  to assign less importance to  $N_{\text{dep}}$  in the current time step, while accounting for a higher impact of  $N_{\text{dep}}$  accumulated in past years. Following this logic, we calculated accumulated  $N_{\text{dep}}$  to account for the total amount of nitrogen that has been deposited over a specific period (here the window size is set as ten years):

$$\text{Accumulated } N_{\text{dep}_t} = \sum_{i=0}^9 N_{\text{dep}_{t-i}} \times \text{Weight}_i \quad (2)$$

Where  $t$  denotes year  $t$ , and  $\text{Weight}_i$  is the weight assigned to the  $N_{\text{dep}}$  value at time step  $t-i$  following the formula:

$$\text{Weight}_i = e^{-\alpha \cdot (9-i)} \quad (3)$$

Where  $\alpha$  is the decay rate parameter controlling the rate of decay and is set as a constant value of 0.1.  $i$  ranges from 0 to 9, representing the past ten time steps within the window. This characteristic of the exponential decay allows for a balance between considering recent values and incorporating historical data in the accumulation process.

Using the same sets of tests as Table S4, but only changing the method of accumulated  $N_{\text{dep}}$  (Equation 2, 3), the results shown in Table S5 are different.

The low  $R^2$  in M4 in all three tests highlights the inadequacy of solely using accumulated  $N_{\text{dep}}$  as a predictor to explain the variance of  $\text{GPP}_{\text{residual}}$ . However, unlike the  $\eta$  in **Test #1** and **Test #2**,  $\eta$  in **Test #3** shows negative sign (Table S5). This discrepancy can be attributed to the declining trend of  $N_{\text{dep}}$  near most EC sites (Figure S10a). This decline aligns with reported trends in  $N_{\text{dep}}$  in Europe and the eastern U.S. (Ackerman et al., 2019; Gilliam et al., 2019; Nopmongcol et al., 2019; Schmitz et al., 2019). The declining trend of annual  $N_{\text{dep}}$  will also lead to declining accumulated  $N_{\text{dep}}$  calculated by equations 2 and 3, as they consider the accumulated  $N_{\text{dep}}$  over the past ten years. It's worth noting that the 166 synthetic sites in QUINCY are randomly distributed across the globe, thus exhibiting both positive and negative trend in  $N_{\text{dep}}$ .

Although the estimation of  $\beta$  by M2 in **Test #3** decreases after incorporating  $N_{\text{dep}}$  into the linear regression model (Table S5), the negative sign of  $\eta$  suggests that it is likely a statistical spurious attribution, primarily due to the declining trend in  $N_{\text{dep}}$ . Experiments show that the positive effect of N addition on productivity persist for a long time even after ceasing the N load (Hrevušová et al., 2009; Meng et al., 2023). The duration for which the benefits of N retention persist after

adding it to the ecosystem is closely linked to the balance between N input from biological nitrogen fixation and  $N_{\text{dep}}$ , and N loss through processes such as leaching and denitrification (Sokolov et al., 2008; Zaehle & Dalmonech, 2011; Davies-Barnard et al., 2020). Uncertainties in the terrestrial N cycling such as the rate of denitrification, pose challenges in understanding the resident time of N, as various factors (e.g., climate, vegetation type, soil texture, pre-disturbance nutrient levels) can influence the processes (Galloway et al., 2004; Sokolov et al., 2008; Davies-Barnard et al., 2020). Given these uncertainties and the scarcity of global data on terrestrial N cycling (Zaehle & Dalmonech, 2011), it is hard to conclude how to integrate  $N_{\text{dep}}$  to account for the cumulative effect of  $N_{\text{dep}}$  on vegetation productivity.

**Table S5.** Linear regression models of the  $GPP_{\text{residual}}$  used to test the cumulative effect of  $N_{\text{dep}}$  calculated by equation 2 and 3

<b>Test #1 <math>GPP_{\text{residual}}</math> modeled by QUINCY</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> ( $\text{gC m}^{-2} \text{yr}^{-1} \text{ppm}^{-1}$ )	<b>Median <math>\eta</math></b> ( $\text{KgC KgN}^{-1}$ )	<b>Median trend</b> ( $\text{gC m}^{-2} \text{yr}^{-2}$ )	<b>Mean <math>R^2</math></b>
<b>M1:</b> $GPP_{\text{residual}} \sim \text{CO}_2$	5.78	-	-	0.718
<b>M2:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + N_{\text{dep}}$	5.73	-1.40	-	0.724
<b>M3:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + \text{years}$	3.95	-	3.37	0.728
<b>M4:</b> $GPP_{\text{residual}} \sim N_{\text{dep}}$	-	44.75	-	0.328
<b>M5:</b> $GPP_{\text{residual}} \sim \text{years}$	-	-	12.14	0.717
<b>Test #2 <math>GPP_{\text{residual}}</math> derived by the <i>GPP residual method</i> in QUINCY simulation</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> ( $\text{gC m}^{-2} \text{yr}^{-1} \text{ppm}^{-1}$ )	<b>Median <math>\eta</math></b> ( $\text{KgC KgN}^{-1}$ )	<b>Median trend</b> ( $\text{gC m}^{-2} \text{yr}^{-2}$ )	<b>Mean <math>R^2</math></b>
<b>M1:</b> $GPP_{\text{residual}} \sim \text{CO}_2$	5.81	-	-	0.355
<b>M2:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + N_{\text{dep}}$	5.67	1.25	-	0.366
<b>M3:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + \text{years}$	2.68	-	6.72	0.369
<b>M4:</b> $GPP_{\text{residual}} \sim N_{\text{dep}}$	-	47.40	-	0.168
<b>M5:</b> $GPP_{\text{residual}} \sim \text{years}$	-	-	12.22	0.369
<b>Test #3 <math>GPP_{\text{residual}}</math> derived by the <i>GPP residual method</i> in EC dataset</b>				
<b>Linear regression models</b>	<b>Median <math>\beta</math></b> ( $\text{gC m}^{-2} \text{yr}^{-1} \text{ppm}^{-1}$ )	<b>Median <math>\eta</math></b> ( $\text{KgC KgN}^{-1}$ )	<b>Median trend</b> ( $\text{gC m}^{-2} \text{yr}^{-2}$ )	<b>Mean <math>R^2</math></b>
<b>M1:</b> $GPP_{\text{residual}} \sim \text{CO}_2$	3.18	-	-	0.100
<b>M2:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + N_{\text{dep}}$	2.01	-18.59	-	0.143
<b>M3:</b> $GPP_{\text{residual}} \sim \text{CO}_2 + \text{years}$	-4.07	-	22.24	0.166
<b>M4:</b> $GPP_{\text{residual}} \sim N_{\text{dep}}$	-	-43.79	-	0.071
<b>M5:</b> $GPP_{\text{residual}} \sim \text{years}$	-	-	13.84	0.142

Isolating the long-term effects, including  $N_{\text{dep}}$  and  $\text{CO}_2$ , poses great challenge. In our tests, we accumulated  $N_{\text{dep}}$  using two different methods: i) cumulative way (Equation 1); ii) accumulating by applying an exponential decay with sliding window (Equation 2, 3). The divergent results highlight the importance of future research in selecting appropriate time-scales, and employing causal inference methods to more effectively disentangle the long-term effects on GPP in ecosystems with tightly coupled carbon-nitrogen-climate interactions.

References:

- Ackerman, D., Millet, D. B., & Chen, X. (2019). Global Estimates of Inorganic Nitrogen Deposition Across Four Decades. *Global Biogeochemical Cycles*, *33*(1), 100–107. <https://doi.org/10.1029/2018GB005990>
- Davies-Barnard, T., Meyerholt, J., Zaehle, S., Friedlingstein, P., Brovkin, V., Fan, Y., Fisher, R. A., Jones, C. D., Lee, H., Peano, D., Smith, B., Wårlind, D., & Wiltshire, A. J. (2020). Nitrogen cycling in CMIP6 land surface models: Progress and limitations. *Biogeosciences*, *17*(20), 5129–5148. <https://doi.org/10.5194/bg-17-5129-2020>
- Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P., Asner, G. P., Cleveland, C. C., Green, P. A., Holland, E. A., Karl, D. M., Michaels, A. F., Porter, J. H., Townsend, A. R., & Vöosmarty, C. J. (2004). Nitrogen Cycles: Past, Present, and Future. *Biogeochemistry*, *70*(2), 153–226. <https://doi.org/10.1007/s10533-004-0370-0>
- Gilliam, F. S., Burns, D. A., Driscoll, C. T., Frey, S. D., Lovett, G. M., & Watmough, S. A. (2019). Decreased atmospheric nitrogen deposition in eastern North America: Predicted responses of forest ecosystems. *Environmental Pollution*, *244*, 560–574. <https://doi.org/10.1016/j.envpol.2018.09.135>
- Gruber, N., & Galloway, J. N. (2008). An Earth-system perspective of the global nitrogen cycle. *Nature*, *451*(7176), 293–296. <https://doi.org/10.1038/nature06592>
- Hrevašová, Z., Hejman, M., Pavlu, V. V., Hakl, J., Klaudivová, M., & Mrkvička, J. (2009). Long-term dynamics of biomass production, soil chemical properties and plant species composition of alluvial grassland after the cessation of fertilizer application in the Czech Republic. *Agriculture, Ecosystems & Environment*, *130*(3), 123–130. <https://doi.org/10.1016/j.agee.2008.12.008>
- Meng, Y., Li, T., Liu, H., Li, S., Xu, Z., & Jiang, Y. (2023). Legacy effects of nitrogen deposition and increased precipitation on plant productivity in a semi-arid grassland. *Plant and Soil*, *491*(1), 69–84. <https://doi.org/10.1007/s11104-022-05550-x>
- Nopmongcol, U., Beardsley, R., Kumar, N., Knipping, E., & Yarwood, G. (2019). Changes in United States deposition of nitrogen and sulfur compounds over five decades from 1970 to 2020. *Atmospheric Environment*, *209*, 144–151. <https://doi.org/10.1016/j.atmosenv.2019.04.018>
- Schmitz, A., Sanders, T. G. M., Bolte, A., Bussotti, F., Dirnböck, T., Johnson, J., Peñuelas, J., Pollastrini, M., Prescher, A.-K., Sardans, J., Verstraeten, A., & de Vries, W. (2019). Responses of forest ecosystems in Europe to decreasing nitrogen deposition. *Environmental Pollution*, *244*, 980–994. <https://doi.org/10.1016/j.envpol.2018.09.101>
- Sokolov, A. P., Kicklighter, D. W., Melillo, J. M., Felzer, B. S., Schlosser, C. A., & Cronin, T. W. (2008). Consequences of Considering Carbon–Nitrogen Interactions on the Feedbacks between Climate and the Terrestrial Carbon Cycle. *Journal of Climate*, *21*(15), 3776–3796. <https://doi.org/10.1175/2008JCLI2038.1>
- Zaehle, S., & Dalmonech, D. (2011). Carbon–nitrogen interactions on land at global scales: Current understanding in modelling climate biosphere feedbacks. *Current Opinion in Environmental Sustainability*, *3*(5), 311–320. <https://doi.org/10.1016/j.cosust.2011.08.008>