
Supplementary information

Terrestrial photosynthesis inferred from plant carbonyl sulfide uptake

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Supplementary Materials for

Terrestrial Photosynthesis Inferred from Plant Carbonyl Sulfide Uptake

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Supplementary Text S1. Potential uncertainties in the g_{mes} model

Our g_{mes} was assumed to be tightly linked to M_a following a decreasing power law, a broad pattern drawn from a meta-analysis across diverse plant species with different foliage structures¹. However, the assumed g_{mes} - M_a relationship may not apply for certain species, e.g., Australian sclerophylls^{2,3}. This may have led to the smaller or even negative impact of g_{mes} on OCS fluxes of C3 arctic grass in high northern latitudes (Fig. 2). Future studies should refine the g_{mes} - M_a relationship by extending to broader plant species to improve its global generalizability.

Moreover, our g_{mes} model adopts the temperature response function from Bernacchi et al.⁴ and water stress formulation from Oleson et al.⁵, respectively, assuming they can capture the first-order impacts of environmental stress. However, response and future acclimation of g_{mes} to these environmental factors are highly complex, varying widely across plant species, temperature ranges, growth conditions, etc.⁶⁻⁸. These complexities were not fully represented by our model due to limited measurements for model parameterization, which may lead to uncertainties in the simulated g_{mes} impact on global carbon and water fluxes⁹. Future studies are urgently needed to understand the mechanistic drivers of g_{mes} as well as the acclimation of g_{mes} to future climate changes and develop formulations to represent such processes.

Supplementary Text S2. Impact of LRU treatments (constant vs. PAR dependent) on GPP estimates

Different LRU treatments can lead to large differences in the OCS-inferred GPP. A constant LRU value (1.61 for C3 and 1.16 for C4, the median values reported previously¹⁰ and often adopted at regional/global scales¹¹) would lead to a global annual mean of $GPP_{\text{OCS_LRU_constant}}$ up

to 182 PgC yr⁻¹ with explicit g_{mes} (Fig. 4a and Table S1). This is because under low light conditions (e.g., in the early morning, late afternoon), the actual LRU (which decreases with light) should be much larger than the constant value of 1.61, thus GPP based on this constant LRU would be overestimated. This is especially the case at nighttime when there is an active plant OCS and thus an incorrect non-zero GPP estimate if using a constant LRU value, while in reality there should be no GPP at nighttime (Fig. 3). In contrast, with an implicit g_{mes} treatment, the global mean of $GPP_{OCS_LRU_constant}$ is much smaller, i.e., 131 PgC yr⁻¹, mainly due to the weaker plant OCS flux by g_{mes} -implicit simulation than by g_{mes} -explicit simulation (figs. S18-S20). Note the $GPP_{OCS_LRU_constant}$ with an implicit g_{mes} treatment is also weaker than $GPP_{OCS_LRU_PAR}$ (figs. S18-S20), particularly under high light conditions, when the value of 1.61 is highly likely to be at the higher end of the LRU range^{12,13}. Recent studies based on the optimality theory reported much lower LRU values under light-saturated (0.68 to 1.58)¹⁴, of which LRU at FI-Hyy (1.07 on average for PAR > 700 $\mu\text{mol m}^{-2}\text{s}^{-1}$) falls in between. This likely higher value of constant LRU leads to an underestimation of GPP under high light and can explain the much smaller $GPP_{OCS_LRU_constant}$ (with implicit g_{mes}) than $GPP_{OCS_LRU_PAR}$ (figs. S18-S20). Thus, considering the variability of LRU under different environmental conditions is vital for an accurate GPP estimation if inferred from OCS.

Supplementary Text S3. Shared uncertainties by existing satellite optical remote sensing driven GPP products

The generally lower estimates of global annual mean GPP from existing GPP products driven by satellite optical data likely reflect their shared biases or uncertainties, regardless of whether these estimates are based on the LUE approach, machine learning, or linear scaling of SIF. These biases and uncertainties may include: 1) spatial representativeness, as satellite SIF- and VI-based global GPP estimates are extrapolated from relationships calibrated at a limited number of flux towers unevenly distributed across ecosystem types and hydroclimatic regimes¹⁵ 2) bias in *in-situ* GPP due to imperfect NEE partitioning algorithms¹⁶⁻¹⁸, and 3) bias in global remote sensing products as input, including SIF, fPAR, LAI, VIs, and/or meteorological data, particularly in the tropics due to cloud contamination and saturation of VIs for dense vegetation^{15,19,20}. Additionally, the divergent global and regional GPP patterns among these products can be

attributed to different meteorological forcings, different algorithms to establish relationships between GPP and input variables, with or without considerations of CO₂ fertilization effect, and/or calibration against *in-situ* flux data with varying temporal coverages, etc^{19,21,23}.

Supplementary Text 4. Sensitivity analysis on the impact of ENSO on the contemporary GPP estimates

Note that our OCS-inferred GPP estimate is for the period from 2000 to 2010 which experienced no major El Niño/Southern Oscillation (ENSO) events. As ENSO could strongly impact GPP^{24,25}, one may expect a greater GPP during our study period than if an El Niño event had existed, especially in the pan- tropics²⁵. To evaluate the possibility that our higher global GPP estimates might be an artifact of the lack of ENSO events, we conducted sensitivity tests and compared the GPP simulations of the 1997–1998 El Niño and the 1998–1999 La Niña events (fig. S12). Though considerable GPP reduction during El Niño events appeared in tropical regions, little ENSO impact was found on the global estimates (the global mean GPP difference between El Niño and La Niña events is less than 1 PgC yr⁻¹), indicating the robustness of our OCS-inferred contemporary GPP estimates.

Supplementary Text S5. Technical procedures of Monte Carlo simulations for quantifying impact of cross-PFT variations in LRU and LRU-PAR dependency on GPP estimates

Detailed steps of our Monte Carlo simulations are given as follows:

1. We screen measurements from Stimler et al.²⁶ (their Table II, which contains LRU values for different species at 3 different light levels) that satisfied an expected LRU-PAR relationship, i.e., LRU decreases with increasing light intensity (fig. S15a).
2. Based on the selected measurements in step (1), we calculate the standard deviation (S.D.) of LRUs at the three light levels, and construct a relationship between S.D. of LRU and 1/PAR, assuming they also follow a linear relationship (fig. S15b).
3. For each C3 PFT, do steps (a) to (c).
 - a. Randomly sample 10% of the *in-situ* measurements of LRU and PAR pairs from Kooijmans et al.²⁷.

- b. For each sampled LRU in step (a), we calculate its corresponding S.D. under the same PAR level using the linear relationship established in (2), and add random errors drawn within $\pm 1.96 \times \text{S.D.}$ (corresponding to the 95% confidence interval, assuming the species-specific LRU follows a normal distribution at each PAR level) (fig. S15b).
 - c. Fit a new LRU vs 1/PAR relationship using the paired LRU (with random error added) and PAR samples in step (b) (fig. S15c).
4. With LRU vs 1/PAR formulations established separately for each C3 PFT, we calculate the corresponding GPP from OCS fluxes across the globe.
5. We repeat steps (3) to (4) 100 times, and compute the uncertainty range ($\pm 1 \text{ S.D.}$) of GPP arising from cross-species/PFT LRU variability.

Supplementary Text S6. Model-data comparison of our CLM5-simulated GPP and site measurements at four tropical sites

We conducted CLM5 simulation of GPP (from OCS-inferred and FvCB) at the grid levels within which the four Amazon flux tower sites are located (Table S2). A point simulation at these sites is technically feasible, but requires substantial site-specific info (e.g., OCS concentration, LRU measurements), which we do not have, especially for OCS modeling. We were able to conduct point simulations of OCS at FI-Hyy and US-Ha1 because site-specific parameters/observations, e.g., LRU (for FI-Hyy), OCS concentration, OCS fluxes, are available. It is crucial to have site-specific info to ensure the agreement between modeled GPP and *in-situ* GPP, especially for these tropical sites with distinct communities of vegetation structural and functional composition. Therefore, we decided to compare our simulations at the grid cell and compare the simulated GPP with *in-situ* GPP at these four sites. The simulation period was determined based on the available GPP data at the four tropical EC sites (Table S2). Another constraint of the ending year of CLM5 simulation is the available forcing data which lasts only until 2014.

Our model-data comparison at the four tropical Amazon sites focusses mainly on the wet- vs. dry-season contrast (fig. S14). This is because that the seasonal shape of GPP in tropical rainforests are driven by distinct leaf phenology/leaf ages^{28,29} that are not yet fully represented by any of the existing carbon cycle models (including CLM)³⁰. Therefore, neither g_{mes} or OCS would

be the panacea to solve the incapability of existing carbon cycle models in reproducing the observed seasonality in tropical rainforests^{31,32}.

Supplementary Table S1. Global budget of GPP (averaged from 2000 to 2010) as directly simulated by CLM5 FvCB (GPP_{CLM5_FvCB}), inferred from plant OCS uptake using the PAR-dependent LRU equation (Eq. 25) ($GPP_{OCS_LRU_PAR}$), and constant LRU value (1.61) ($GPP_{OCS_LRU_constant}$) for C3 species (for C4 species, the LRU was all set as 1.16^{10,33}), with implicit or explicit consideration of g_{mes} . Unit: PgC yr⁻¹.

	GPP_{CLM5_FvCB}	$GPP_{OCS_LRU_PAR}$	$GPP_{OCS_LRU_constant}$
g_{mes}-implicit	120	152	131
g_{mes}-explicit	126	157 (± 8.5)	182

Supplementary Table S2 Site characteristics of four selected tropical sites (following Restrepo-Coupe et al.³⁰) utilized to evaluate OCS-inferred GPP. In-situ GPP at these sites come from Large-Scale Biosphere-Atmosphere Experiment in the Amazon (LBA) (https://daac.ornl.gov/LBA/guides/CD32_Fluxes_Brazil.html).

Tower code	Site (region)	Location (longitude, latitude)	Data period used for validation
K67	Para Western (Santarem) - km 67 Primary Forest Tower	(-54.959, -2.857)	2002-2005
CAX	Para Eastern (Belem) - FLONA Caxiutana	(-51.4536, -1.7483)	2000-2003
K34	Amazonas (Manaus) - ZF2 km 34	(-60.1045, -2.5545)	2000-2006
RJA	Rondonia - Jaru Biological Reserve Tower B	(-61.9331, -10.078)	2000-2002

Supplementary Table S3. Characteristics of the two sites with continuous multi-year measurements of OCS fluxes

Site code	Location	PFT	Time period	Reference
US-Ha1 ^a	Harvard forest, USA	Deciduous broadleaf forest	Jan to Nov, 2012 to 2013	27
FI-Hyy ^b	Hyytiälä, Finland	Boreal forest	Apr to Oct, 2013 to 2017	34

a: GPP is partitioned from *in-situ* NEE measurements using daytime³⁵, nighttime³⁶, and isotope-based algorithms¹⁸. For isotope-based partitioning, data is only available from daily 5:00 to 18:00.

b: GPP is partitioned from *in-situ* NEE measurements using nighttime-based approach³⁶.

Supplementary Table S4. PFT-specific values of key parameters in modeling soil OCS fluxes, adopted from Meredith et al.^{37,38}

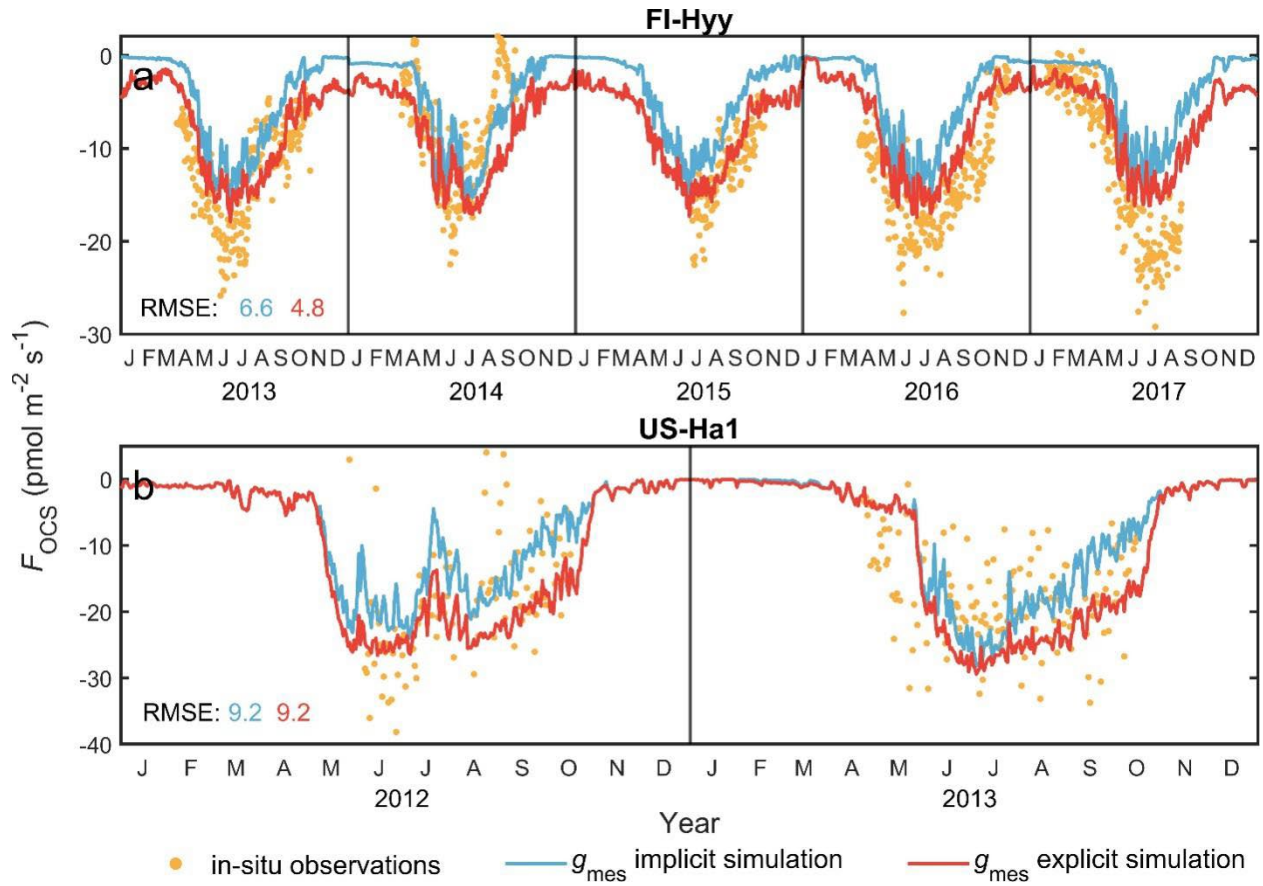
PFT	f_{CA} (Eq. 18)	j (Eq. 23)	m (Eq. 23)
grass	45000	2.20	0.096
evergreen forest	32000	4.86	0.101
deciduous forest	32000	4.94	0.107
agriculture	6500	9.59	0.104
desert/bare soil	13000	5.60	0.050

Supplementary Table S5. Descriptions on three types of GPP estimates inferred from OCS (but using different LRU values) or simulated from CLM5 with FvCB model.

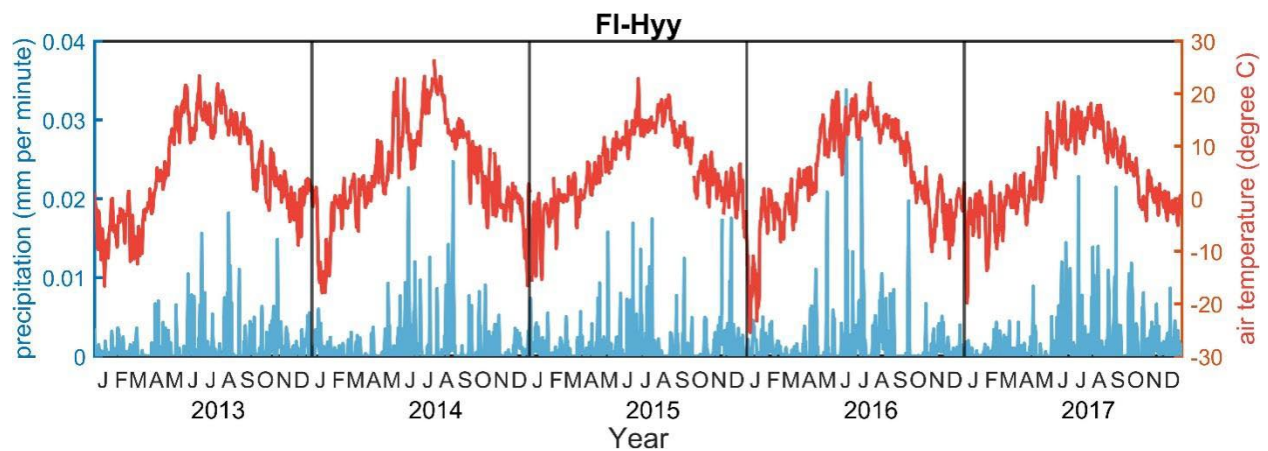
GPP estimate	Description (C3 only)
$GPP_{OCS_LRU_PAR}$	Inferred from simulated OCS LRU=607.2623/PAR+0.5705
$GPP_{OCS_LRU_constant}$	plant uptake with different LRU values for C3 species LRU = 1.61
GPP_{CLM5_FvCB}	Simulated from CLM5 with FvCB model

Supplementary Table S6. Information on model simulation scenario

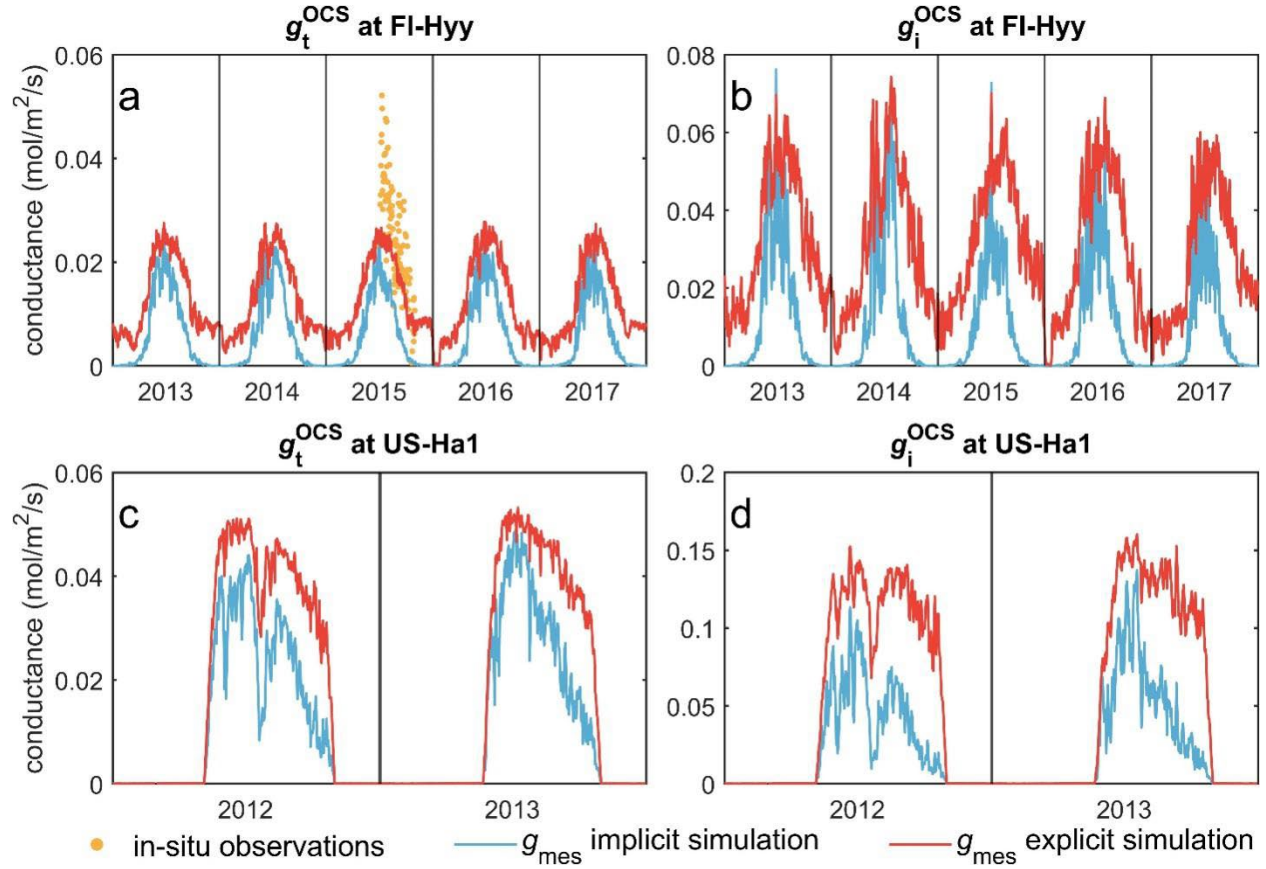
Simulation	Calculation of g_t^{OCS}	Comparison with S2
S1	$g_t^{OCS} = \frac{1}{\frac{1}{g_b^{OCS}} + \frac{1}{g_s^{OCS}} + \frac{1}{g_i^{OCS}}}; g_i^{OCS} = \alpha \cdot V_{cmax} \cdot f_w(\theta) \cdot \left(\frac{p}{p_0}\right) \cdot \left(\frac{T_{can}}{T_0}\right)$	Effect of mechanistic consideration of mesophyll diffusion on OCS simulation
S2	$g_t^{OCS} = \frac{1}{\frac{1}{g_b^{OCS}} + \frac{1}{g_s^{OCS}} + \frac{1}{g_{mes}^{OCS}} + \frac{1}{g_{CA}^{OCS}}}; g_{mes}^{OCS} \text{ by Sun et al.}^{39}; g_{CA}^{OCS}=0.055$	
S3	$g_t^{OCS} = \frac{1}{\frac{1}{g_b^{OCS}} + \frac{1}{g_s^{OCS}} + \frac{1}{g_{CA}^{OCS}}}; g_{CA}^{OCS}=0.055$	Effect of mesophyll diffusion on OCS simulation
S4	$g_t^{OCS} = \frac{1}{\frac{1}{g_b^{OCS}} + \frac{1}{g_s^{OCS}} + \frac{1}{g_{mes}^{OCS}} + \frac{1}{g_{CA}^{OCS}}}; g_{mes}^{OCS} \text{ by Sun et al.}^{39}; g_{CA}^{OCS}=f(T)$	Impact of g_{CA}^{OCS} parameterization on OCS simulation



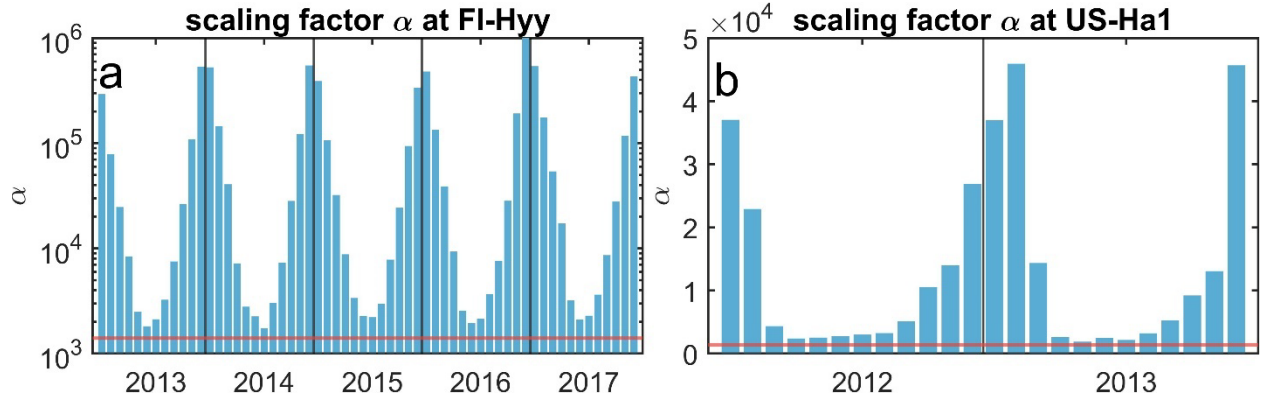
Supplementary Figure S1. Time series of terrestrial ecosystem OCS fluxes (F_{OCS}) simulated by implicit and explicit representations of mesophyll conductance (g_{mes}) and *in-situ* ecosystem-scale measurements at (a) Hyytiälä Forest, Finland (FI-Hyy) and (b) Harvard Forest, Massachusetts, USA (US-Ha1). Negative F_{OCS} denotes net OCS uptake. RMSE of g_{mes} -implicit and g_{mes} -explicit simulations referenced to *in-situ* measurements are given in blue and red texts respectively.



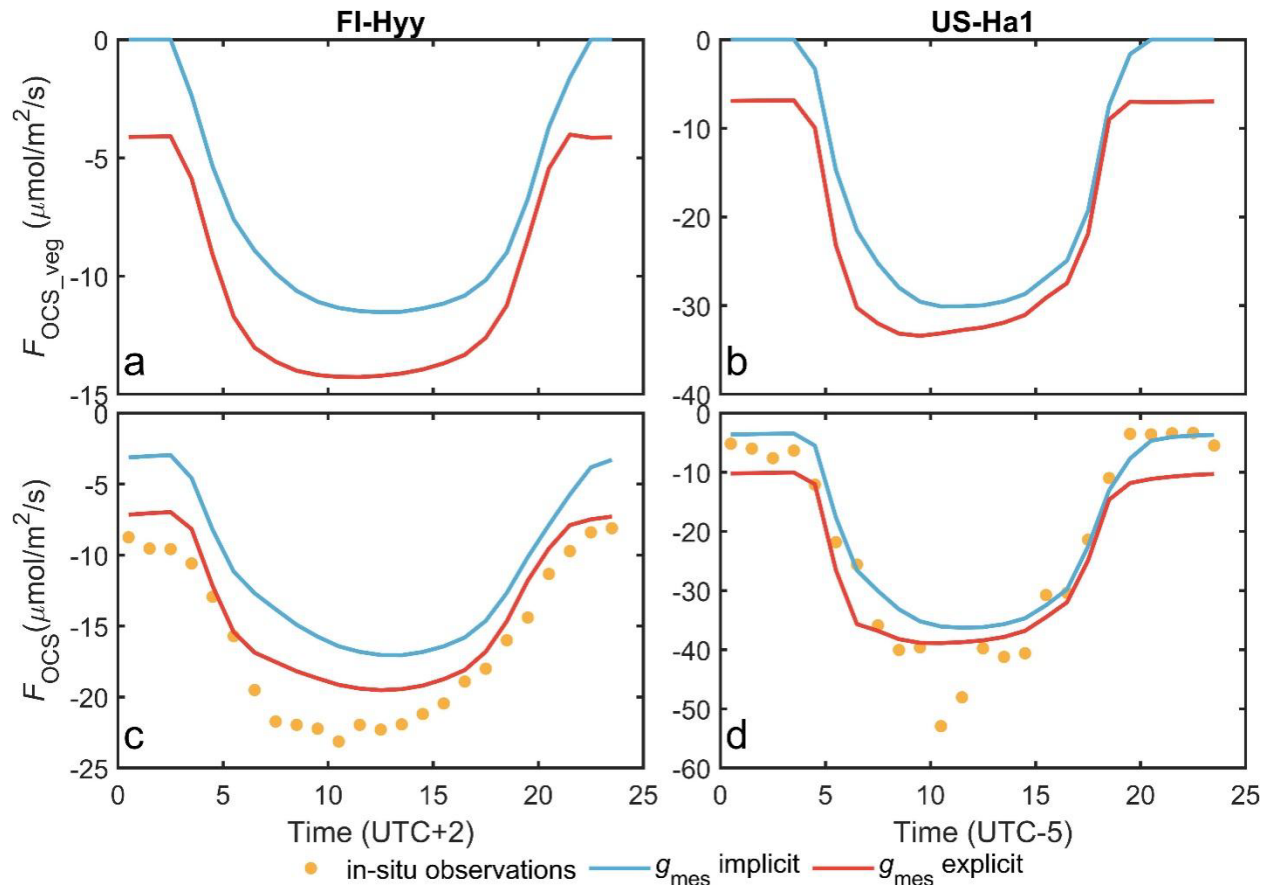
Supplementary Figure S2. Seasonal dynamics of precipitation (in blue) and air temperature (in red) measured at FI-Hyy.



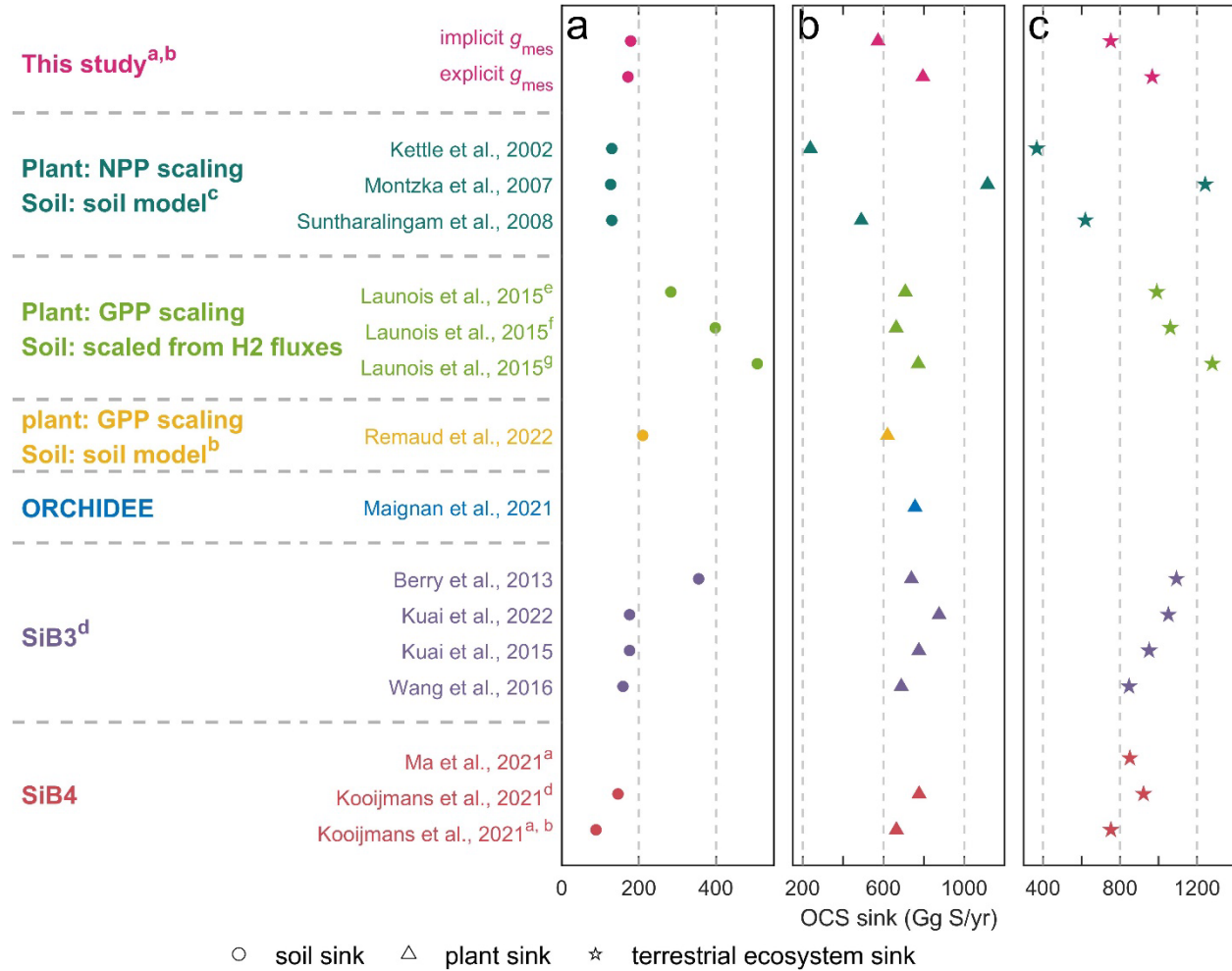
Supplementary Figure S3. Comparison of g_t^{OCS} and g_i^{OCS} (defined as $1/(1/g_{mes}^{OCS} + 1/g_{CA}^{OCS})$, Methods) simulated by g_{mes} -implicit and g_{mes} -explicit across at FI-Hyy and US-Ha1. In-situ g_t^{OCS} was calculated as plant OCS fluxes (ecosystem OCS flux minus the OCS soil flux) divided by the measured ambient OCS concentration.



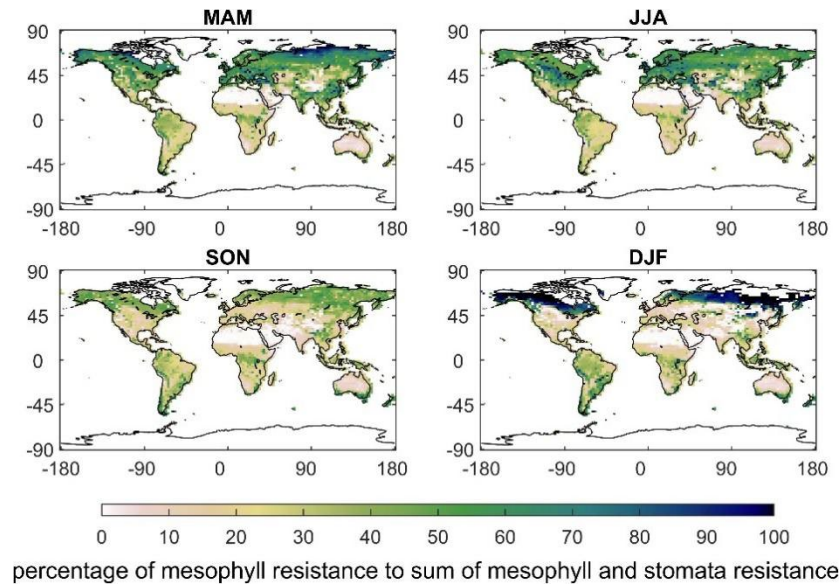
Supplementary Figure S4. Monthly values of the scaling factor α in g_{mes} -implicit simulation required to match the simulated F_{OCS} by the g_{mes} -explicit simulation (Methods). Specifically, α is calculated as: $\alpha = 1400 * g_i^{\text{OCS}}(g_{\text{mes}}\text{-explicit}) / g_i^{\text{OCS}}(g_{\text{mes}}\text{-implicit})$. The horizontal line is the α value adopted by Berry et al.⁴⁰.



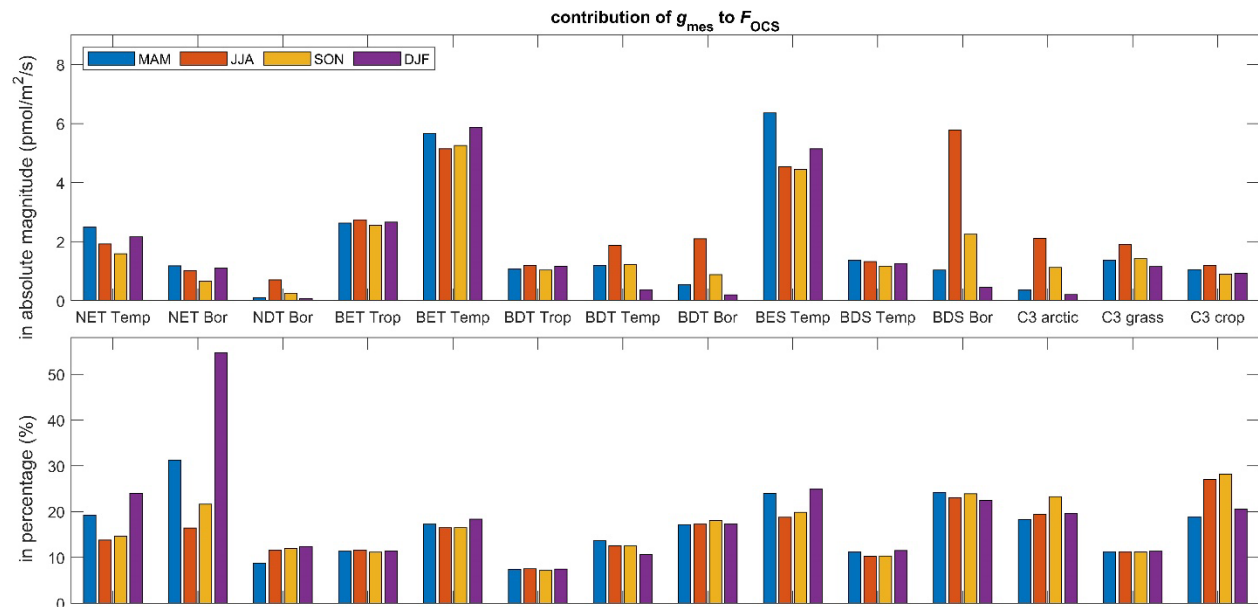
Supplementary Figure S5. Comparison of diurnal cycle of g_{mes} -implicit and g_{mes} -explicit simulated OCS plant flux ($F_{\text{OCS_veg}}$) and OCS ecosystem flux (F_{OCS}) in the growing season (from June to August). In situ observation of F_{OCS} was also plotted as a reference.



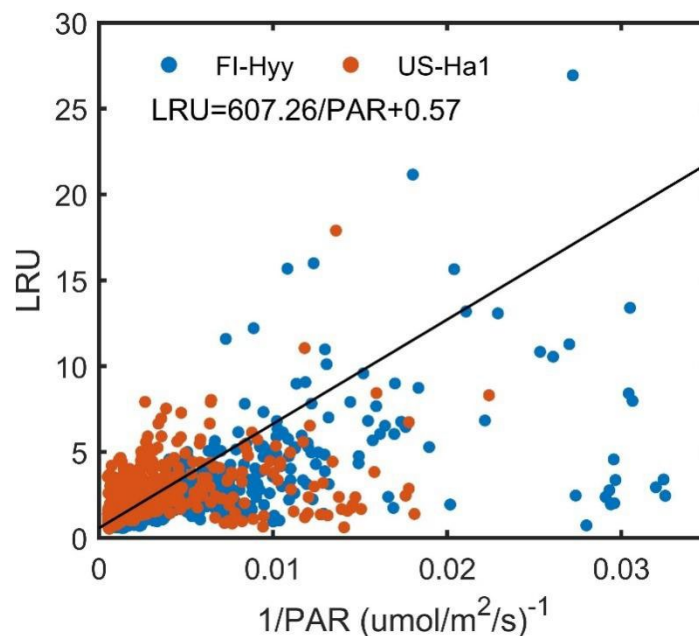
Supplementary Figure S6. Comparison of the global annual OCS (a) soil sink, (b) plant sink and (c) net terrestrial ecosystem sink, estimated in this study (average of 2000 to 2010) and in previous studies. Note a positive OCS sink corresponds to net OCS uptake. Superscripts: a, consideration of monthly and globally varied concentration of atmospheric OCS; b, based on soil OCS model of ref⁴¹; c, based on soil OCS model of ref⁴²; d, soil OCS flux scaled by heterotrophic respiration of CO₂; e, simulations from Organising Carbon and Hydrology In Dynamic Ecosystems (ORCHIDEE) model; f, simulations from Lund-Potsdam-Jena (LPJ) model; g, simulations from Community Land Model (CLM) 4.



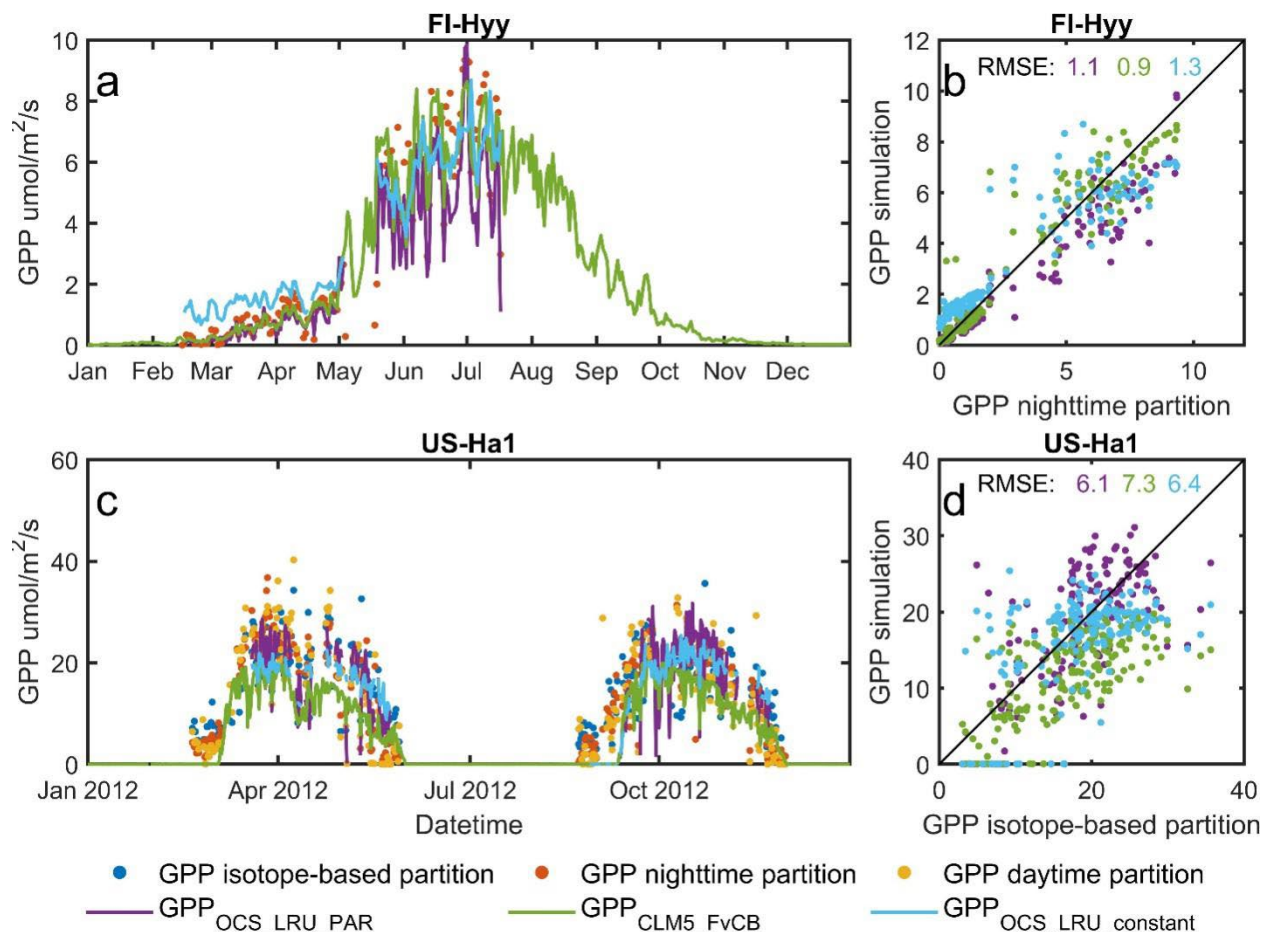
Supplementary Figure S7. The percentage of mesophyll resistance to the sum of mesophyll and stomata resistance (in %). Here we use the resistance term rather than conductance, because the former is additive.



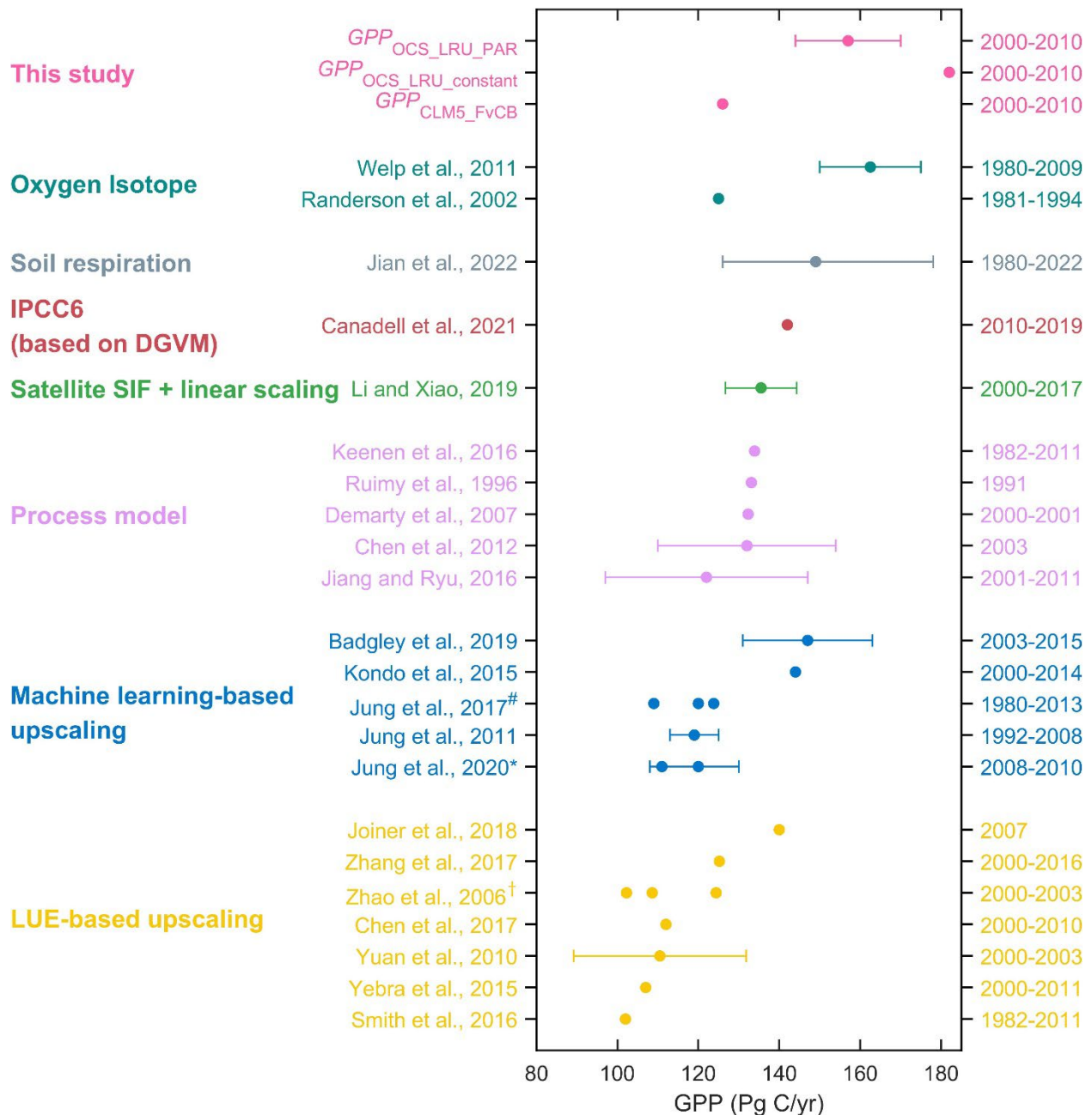
Supplementary Figure S8. Contribution of mesophyll resistance to F_{OCS} by season (a) in absolute magnitude, and (b) in percentage. The contribution was computed as the difference in simulated F_{OCS} between g_{mes} - explicit (S2) and g_{mes} - excluding (S3) experiments (Methods).



Supplementary Figure S9. Relationships between LRU and 1/PAR. The black line shows the empirical equation between LRU and 1/PAR (Eq. 25) fitted with leaf-level measurements in ref²⁷. Blue and red dots display the ecosystem-level LRU ~ 1/PAR relationships at FI-Hyy and US-Ha1 respectively. To derive canopy level LRU, we calculated the ratio of OCS plant flux and GPP, both fluxes normalized by the ambient concentration of the respective gases (OCS and CO₂). Specific to ecosystem-level GPP, it is inferred from the standard daytime partitioning approach³⁵ at HI-Fyy, and from the stable carbon isotope-based partitioning at US-Ha1 (given its better performance thanks to the independent isotopic instrumentation available at this site)¹⁸.

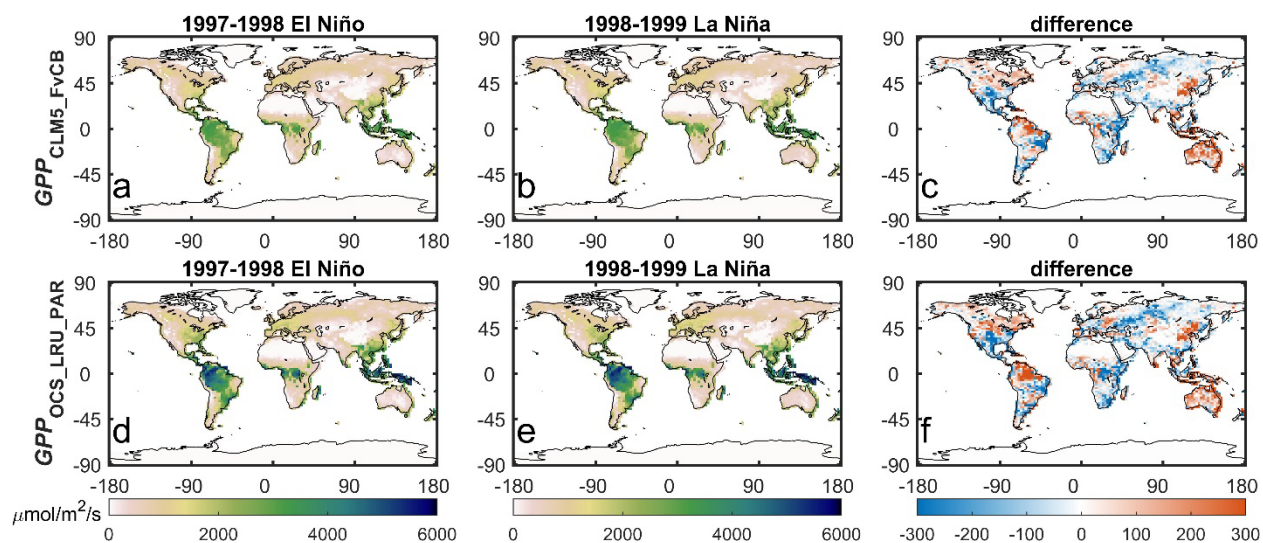


Supplementary Figure S10. Seasonal dynamics of daily GPP from site partitioning estimations (dots, with colors denoting different partitioning method), inferred from OCS simulation (g_{mes} -explicit) with a constant LRU ($\text{GPP}_{\text{OCS_LRU_constant}}$) or PAR-dependent LRU ($\text{GPP}_{\text{OCS_LRU_PAR}}$), and those simulated by CLM5 with FvCB model ($\text{GPP}_{\text{CLM5_FvCB}}$, g_{mes} -explicit) at two sites. Note that, for the US-Ha1 site, daily GPP averaged only from 5:00 to 18:00 local time was displayed here, when partitioning results were available.

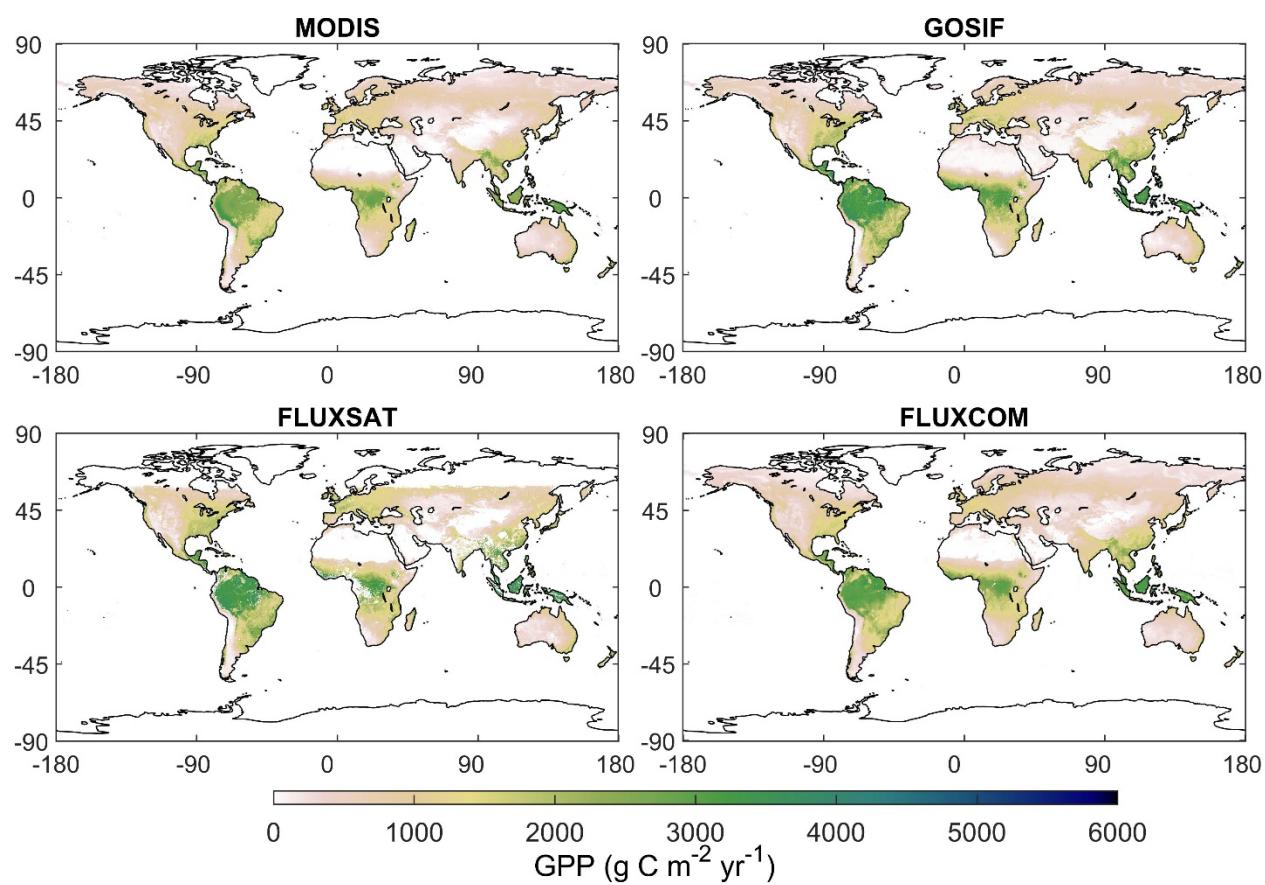


Supplementary Figure S11. Intercomparison of GPP estimates from this study and that from other studies. Colors differentiate the broad categories of methods. References in this figure: ²¹-

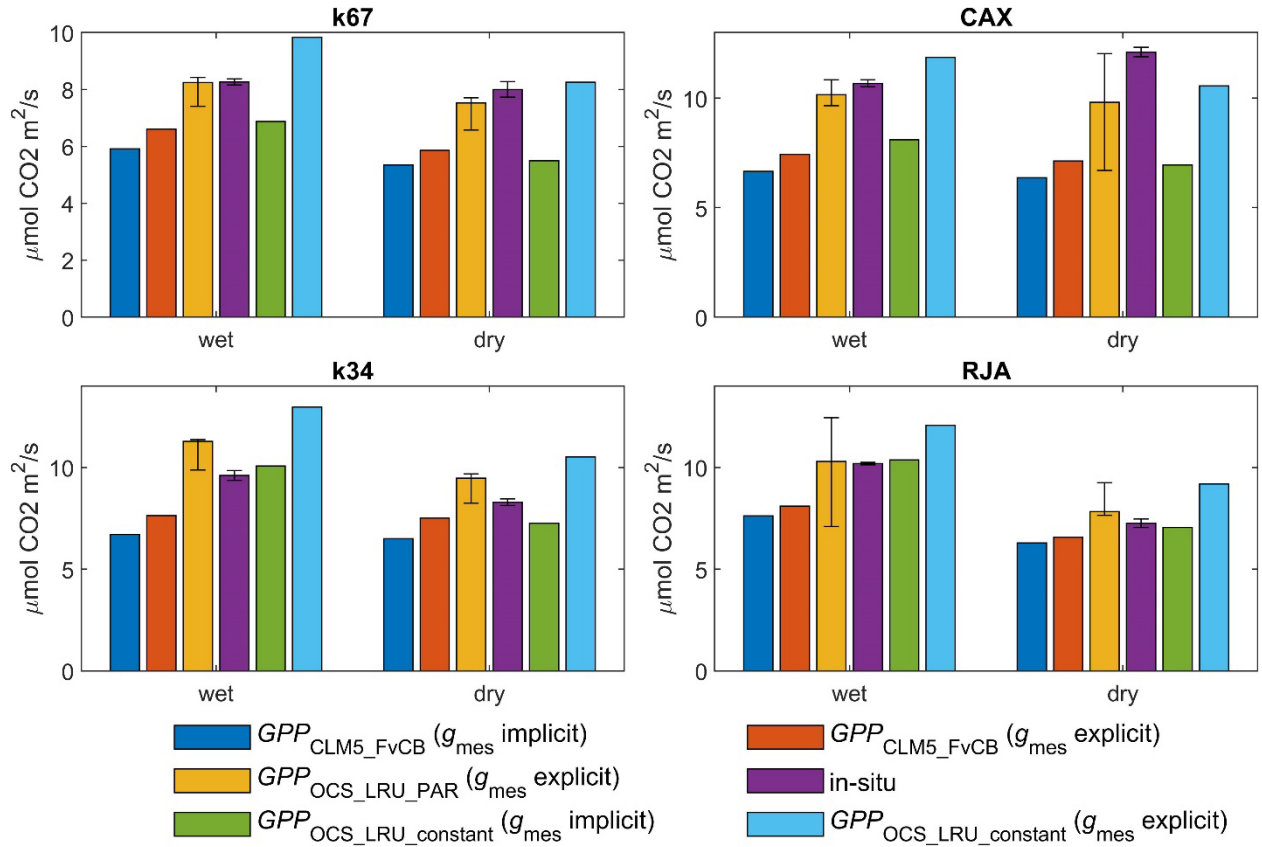
^{24, 43-59} Superscripts: # three GPP products derived from FLUXCOM-ANN (left), FLUXCOM-MARS (middle), and FLUXCOM-RF (right)²². * two GPP products driven by satellite optical data only (left) and by both satellite optical data and meteorological forcing (right)²¹; † three GPP products based on the LUE concept and MODIS optical data but with different meteorological forcings²³.



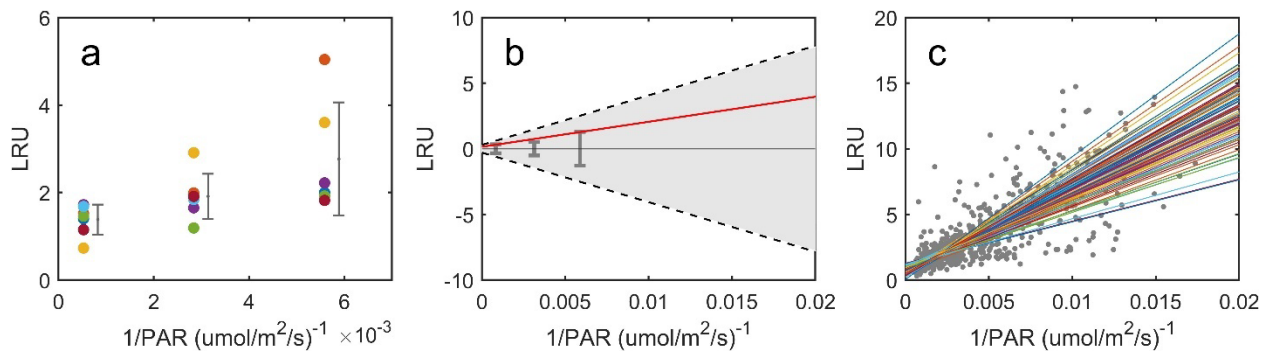
Supplementary Figure S12. GPP estimated by CLM5 with FvCB model (GPP_{CLM5_FvCB}) and that inferred from plant OCS uptake ($GPP_{OCS_LRU_PAR}$) (g_{mes} -explicit) during the 1997-1998 El Niño event (average from May 1997 to May 1998) (a & d), and the 1998-1999 La Niña event (average from May 1998 to May 1999) (b & e), and their difference (La Niña year minus El Niño year) (c & f).



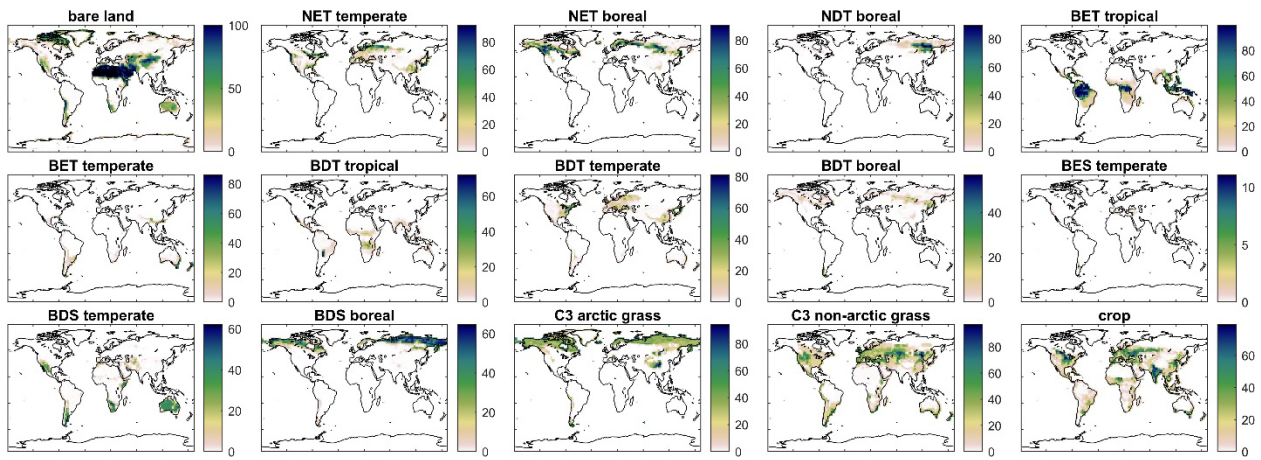
Supplementary Figure S13. GPP maps from four widely used satellite optical remote sensing-driven products in 2010.



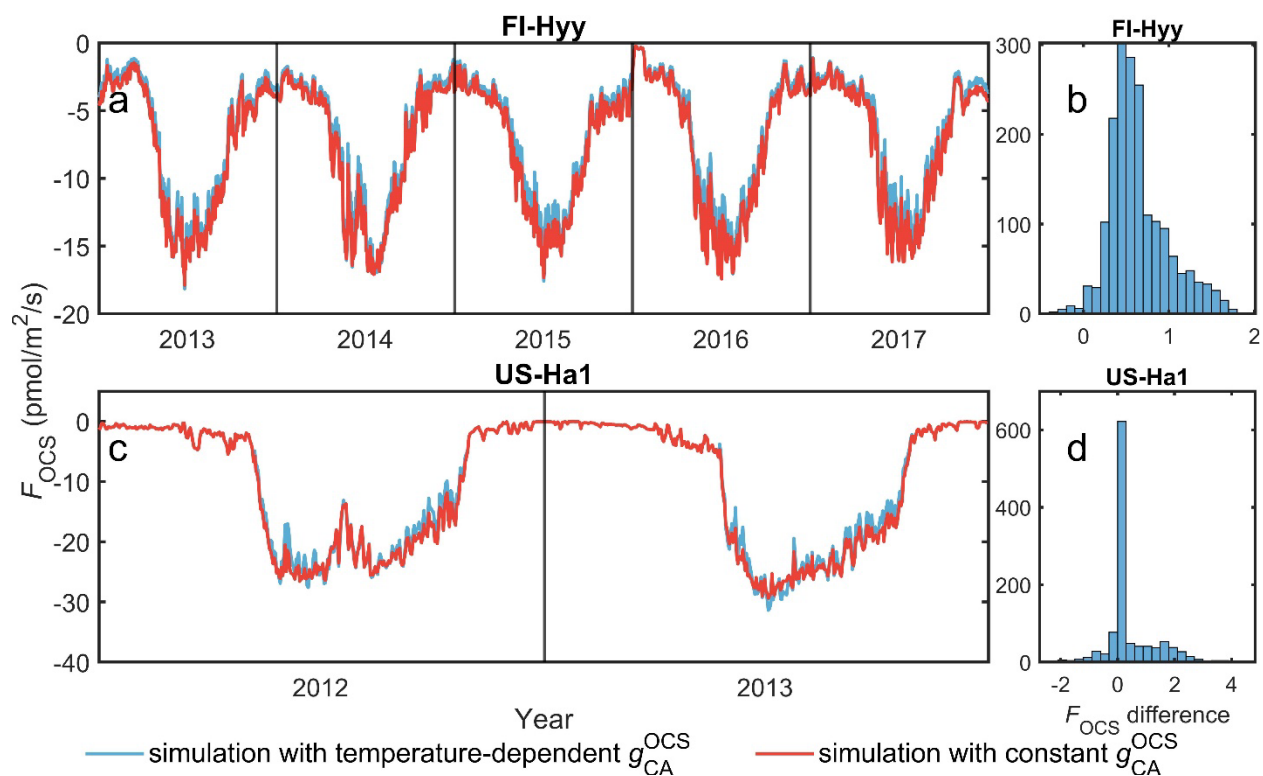
Supplementary Figure S14. Comparison of mean GPP in wet and dry seasons across those simulated directly from CLM5 with FvCB model (GPP_{CLM5_FvCB} , g_{mes} -implicit and g_{mes} -explicit), inferred from plant OCS uptake using the PAR-dependent relationship ($GPP_{OCS_LRU_PAR}$, g_{mes} -explicit) or using a constant LRU ($GPP_{OCS_LRU_constant}$, g_{mes} -implicit and g_{mes} -explicit), with *in-situ* data, at four eddy covariance towers in the tropical Amazon forests. GPP was averaged across years when simulations and *in-situ* data overlap. Dry season for k67 site is defined from July to November, for CAX is from August to November, for k34 is from August to September, for RJA is from May to September; and the other months are wet season.



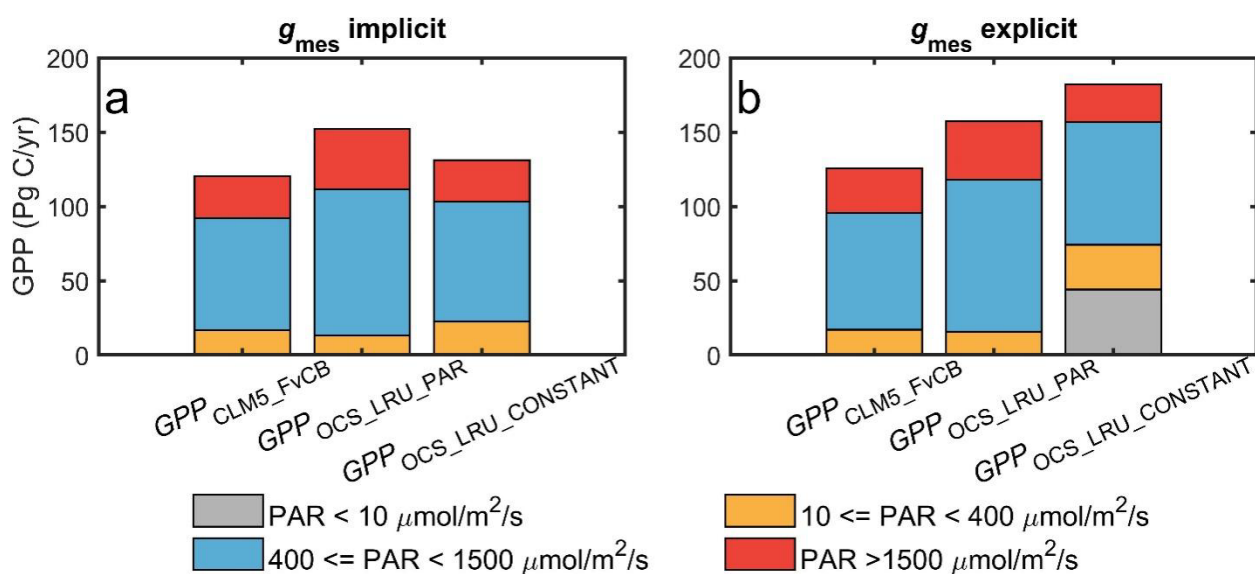
Supplementary Figure S15. a: Selected measurements from Stimler et al.²⁶ (from their Table II) at three different and standardized light levels (i.e., 179, 352, and 1889 $\mu\text{mol m}^{-2}\text{s}^{-1}$) that satisfied an expected LRU-PAR relationship (LRU decreases with increasing light). Points with different colors denote different species. The gray and black bars denote the range of [mean-S.D., mean+S.D.] and [mean-1.96×S.D., mean+1.96×S.D.] (representing the 95% confidence interval of the LRU variability, if we assume the LRU follows a normal distribution). b: Fitted relationship between LRU cross-species S.D. and 1/PAR (represented as red line), and the 95% confidence interval of the LRU uncertainty (represented as the gray shade). c: Illustration of some LRU-PAR relationship fitted using the Monte Carlo method with random uncertainty related to LRU species variability added. The gray dots denote the in-situ measurements of LRU and PAR at the FI-Hyy site²⁷.



Supplementary Figure S16. Percentage coverage (unit: %) of natural PFTs and crop over each global grid set in CLM5. To clarify, CLM5 sets multiple crop functional types, and here we show the sum of percentage coverage of all crop types.

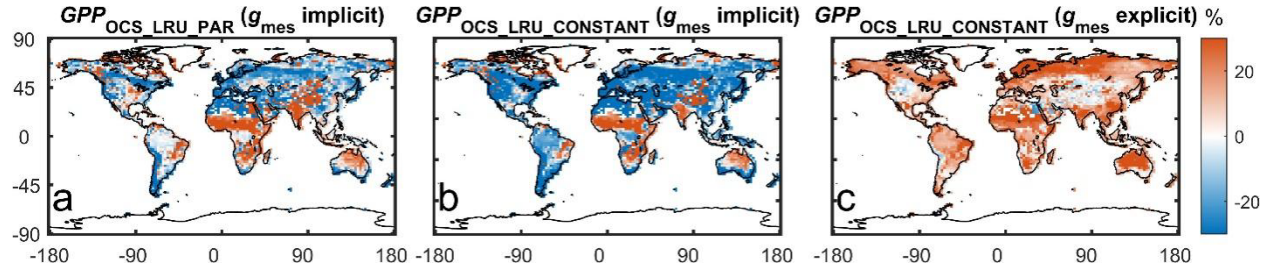


Supplementary Figure S17. a & c: comparison of simulated ecosystem flux (F_{OCS}) with a temperature-dependent g_{CA}^{OCS} (blue line) and those with a constant g_{CA}^{OCS} at leaf scale, same as fig. S1). b & d: histogram of the difference in F_{OCS} by these two simulations.

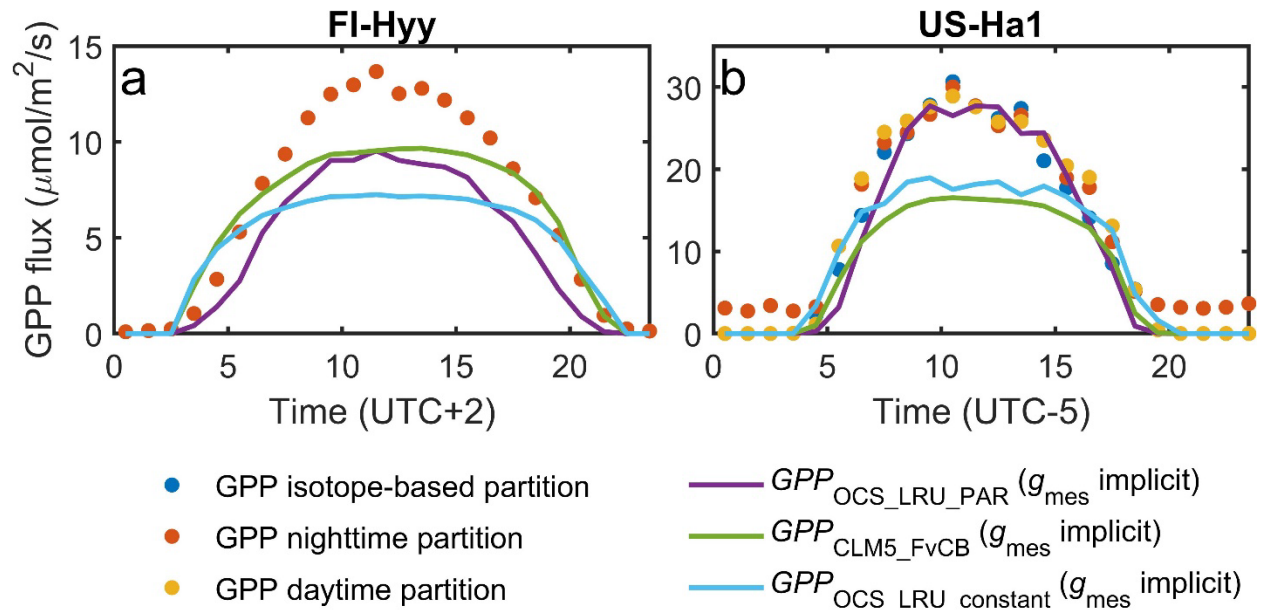


Supplementary Figure S18. Comparison of GPP under different light conditions simulated by CLM5 with FvCB model (GPP_{CLM5_FvCB}), and inferred from plant OCS uptake using PAR-

dependent LRU ($GPP_{OCS_LRU_PAR}$) or constant LRU ($GPP_{OCS_LRU_constant}$) for g_{mes} -implicit (a) and g_{mes} -explicit (b) experiments.



Supplementary Figure S19. Percentage difference of GPP (averaged from 2000 to 2010) simulated by CLM5 with FvCB model (GPP_{CLM5_FvCB} , g_{mes} -implicit) and inferred from plant OCS uptake using constant LRU ($GPP_{OCS_LRU_constant}$, g_{mes} -implicit and g_{mes} -explicit) referenced to the GPP inferred from plant OCS uptake using PAR-dependent LRU ($GPP_{OCS_LRU_PAR}$, g_{mes} -explicit).



Supplementary Figure S20. Evaluation of GPP inferred from plant OCS uptake (g_{mes} -implicit) (with PAR-dependent and constant LRU respectively) and that directly simulated by CLM5 with FvCB model (g_{mes} -implicit) at FI-Hyy and US-Ha1 against GPP partitioned from *in-situ* NEE measurements. Same as Fig. 3a-b, except for g_{mes} -implicit simulations.

Reference

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