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Supporting Information for

**Inferring CO2 fertilization effect based on global monitoring land-atmosphere exchange with a theoretical model**

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**Uncertainty analysis associated with energy imbalance and evaporation contributions**

We quantified the range of uncertainties associated with the energy imbalance of observed data, and intercepted evaporation from the wet surface. Uncertainties by the energy imbalance and intercepted evaporation were estimated using data from selected 14 sites from FLUXNET2015. We quantified a range of the uncertainties for energy imbalance by testing two extreme assumptions (Knauer et al., 2018): 1) all errors were in sensible heat flux and 2) all errors were in latent heat flux. We also quantified uncertainties due to intercepted evaporation, by rejecting data under wet conditions which was defined by 1-day after a rain event (Knauer et al., 2018). The CO2 fertilization was quantified by optimizing the model with corrected heat fluxes or strict criterion for wet conditions as input. Systematic error was estimated based on a Student’s t-test compared with our baseline results. Random errors were estimated as the coefficient of variance in the CO2 fertilization effects among the 14 sites. Uncertainties in CO2 fertilization effect due to energy imbalance and wet surface conditions was small (Fig. S8). The magnitude of the error was less than one tenth the size the magnitude of the CO2 fertilization effects. Energy imbalance induced no systematic error for quantifying the CO2 fertilization effects, with a coefficient of variance: 5.5 % for GPP, 8.1 % for gs, and 3.0 % for iWUE. The CO2 fertilization effects in gs and iWUE systematically decreased 7.9 ± 10.0% and 4.3 ± 6.4%, respectively (p < 0.05), with the criterion for wet surface conditions compared against the baseline. Thus, the systematic errors in gs and iWUE were considered as extending one tail of error bars by the systematic errors in Fig. 1 and 3.

**Model**

*A1. Model*

We developed a sun/shade model (inverse Big-Leaf Model for Eddy Covariance; iBLM-EC version 2.0) with coupled photosynthesis (Farquhar et al., 1980) and stomatal conductance (Ball et al., 1987) sub-models for inverting ecosystem-scale ecophysiological parameters: a maximum carboxylation rate at 25 oC (*Vc*max25), a maximum electron transport rate at 25 oC (*J*max25), and stomatal conductance parameters (*m*bb and *b*bb in Ball et al., 1987). Input variables are wind speed, incoming and reflected photosynthetically active photon flux density (PPFD), air temperature, relative humidity, rainfall, atmospheric pressure, atmospheric carbon dioxide concentration [CO2], net radiation, ground heat flux, friction velocity (*u*\*), sensible and latent heat fluxes, gross primary productivity (GPP), and leaf area index (LAI). Reflected PPFD is calculated based on the radiation transfer model (Ryu et al., 2011).

*A1-1. Photosynthesis model*

Based on the biochemical model (Farquhar et al., 1980), the photosynthetic rate (*A*) is determined as the minimum rate of Rubisco-limited photosynthesis (*A*v) and RuBP-limited photosynthesis (*A*j):

 (A1)

 (A2)

 (A3)

where *R*l is mitochondrial respiration in light, *Vc*max is the maximum carboxylation rate, *J* is the electron transport rate, *Γ*\* is the CO2 compensation point for photosynthesis in the absence of mitochondrial respiration, *p*i is intercellular CO2 partial pressure, *p*o is intercellular O2 partial pressure, and *K*c and *K*o are Michaelis-Menten constants. The variables *Vc*max, *J*, *K*c, *K*o, *Γ*\*, and *R*l change with leaf temperature, and are often parameterized with various types of kinetic functions. We performed simulations with the biochemical model using six different parameterizations (Table A1 in Ueyama et al., 2016) from de Pury & Farquhar (1997), Bernacchi et al. (2001, 2003), Collatz et al. (1991), von Caemmerer et al. (2009), Kosugi et al. (2003), and Kattge & Knorr (2007). While kinetic functions, such as the Arrhenius-type equation, were similar across the six models with only slightly different parameterizations, a model by Kattge & Knorr (2007) considered the acclimation of *Vc*max to growth temperature in the kinetic functions. To introduce a realistic gradual transition from one limitation to another in Eq. A1, we applied an empirical quadratic function proposed by Collatz et al. (1991).

We used a set of kinetic functions and related parameters for the photosynthesis model, for quantifying uncertainties by parameters that were not constrained by data. The parameters included the maximum carboxylation rate(*Vc*max), the maximum electron transport rate(*J*max), Michaelis-Menten constants(*K*c, and *K*o), the CO2 compensation point for photosynthesis in the absence of mitochondrial respiration (*Γ*\*), and mitochondrial respiration in light(*R*l) change with leaf temperature (Table A1 in Ueyama et al., 2016). Basic equations for each model were originally proposed by de Pury & Farquhar (1997), Bernacchi et al. (2001, 2003), Collatz et al. (1991), von Caemmerer et al. (2009), Kosugi et al. (2003), and Kattge & Knorr (2007), but we slightly modified the models and parameterization (Table A1 in Ueyama et al., 2016) from the original ones upon introducing our inversion scheme (shown in section A2).

*Model 1 based on de Pury and Farquhar (1997)*

Temperature dependence of *Vc*max(μmol mol-1), *K*c(Pa), *K*o(Pa), and *R*l (μmol mol-1) are modeled by the Arrhenius function.

 (A4)

where *k*T and k25 are variables (e.g., *Vc*max, *K*c, *K*o, or *R*l) at a given temperature and the reference temperature of 25 oC, respectively. *T*K is leaf temperature in Kelvin, and Ea is the activation energy (J mol-1) (Table A1 in Ueyama et al., 2016), and R is the universal gas constant (8.314 J mol-1 K-1). The temperature dependence of *Γ*\* (Pa) and *J*max (μmol mol-1) are described as follows:

, (A5)

. (A6)

where *J*max25 is the maximum electron transport rate at 25 oC(μmol mol-1),S is an entropy term (J K-1 mol-1), and Ed is the deactivation energy (J mol-1) (Table A1 in Ueyama et al., 2016). The electron transport rate is described as follows:

 (A7)

 (A8)

where θI is the curvature of leaf response of electron transport to irradiance (0.7), *J* is the electron transport rate, *I*le is PPFD effectively absorbed by PSII, *I*l is incoming PPFD, and *f* is the spectral correction factor (0.15).

*Model 2 based on Bernacchi et al. (2001, 2003)*

Temperature dependence of *Vc*max(μmol mol-1), *J*max(μmol mol-1), *K*c(μmol mol-1), *K*o(μmol mol-1), *Γ*\*(μmol mol-1), and *R*l(μmol mol-1) are modeled by the following function (Bernacchi et al., 2001, 2003) (Table A1 in Ueyama et al., 2016).

 (A9)

where c is a scaling constant (Table A1 in Ueyama et al., 2016). The electron transport rate is described as follows:

 (A10)

 (A11)

 (A12)

 (A13)

where *T* is leaf temperature in degree Celsius, ΘPSII is the convexity term for electron transport rates (dimensionless), al is the total leaf absorbance (dimensionless), and ΦPSIImax is the apparent quantum yield of CO2 assimilation (dimensionless).

*Model 3 based on van Caemmerer et al. (2009)*

Temperature dependence of *Vc*max(μmol mol-1), *K*c(Pa), *K*o(Pa) , *Γ*\*(Pa), and *R*l (μmol mol-1) are modeled by the Arrhenius function (Eq. A4) with different parameterization (Table A1 in Ueyama et al., 2016). The electron transport is based on Eq. A6 and A7, where *I*le is defined as:

 (A14)

*Model 4 based on Collatz et al. (2001)*

Temperature dependence of *Vc*max(μmol mol-1), *J*max(μmol mol-1), *K*c(Pa), *K*o(Pa), CO2/O2 specificity ratio (*τ*)(dimensionless), and *R*l (μmol mol-1) are modeled by the *Q*10 function.

 (A15)

where *Q*10 is the relative change in the parameter for a 10 oC change in temperature. In this model, the reduction of *Vc*max under high temperatures is introduced in the following formula:

 (A16)

where *Vc*max0 is *Vc*max calculated using Eq. A15. Temperature inhibition at high temperature is also introduced for *R*l.

 (A17)

where *R*l0 is *R*l calculated using Eq. A15. For calculating the electron transport rate, we used the model (Eq. A6 to A8) by de Pury & Farquhar (1997)*.*

*Model 5 based on Kosugi et al. (2003)*

Temperature dependence of *Vc*max(μmol mol-1) and *J*max(μmol mol-1) is calculated as follows:

 (A18)

The temperature dependence of *K*c(Pa), *K*o(Pa), *τ*, and *R*l (μmol mol-1) are modeled by the Arrhenius function.

 (A19)

For calculating the electron transport rate, we used the model (Eq. A6 to A8) by de Pury & Farquhar (1997)*.*

*Model 6 based on Kattge and Knorr (2007)*

Temperature dependence of *Vc*max(μmol mol-1) and *J*max (μmol mol-1) are calculated using Eq. A6, where *Vc*max is also calculated using same temperature dependence function with different parameters. Temperature dependence of *R*l(μmol mol-1) is calculated by Eq. A9 with a different parameterization (Table A1 in Ueyama et al., 2016). Temperature dependence of *K*c(μmol mol-1), *K*o(μmol mol-1), and *Γ*\*(μmol mol-1) are modeled by Eq. A4. The electron transport rate is described as follows:

 (A20)

 (A21)

*A1-2. Stomatal conductance model*

Based on Ball et al. (1987), stomatal conductance are modelled:

 (A22)

where *g*sw is stomatal conductance to water vapor, *rh*s is relative humidity at the leaf surface (Eq. A32), *c*s is CO2 concentration at the leaf surface, *m*bb is an empirical parameter for a dimensionless slope, and *b*bb is an empirical parameter for the zero intercept when net photosynthetic rate is equal or less than zero. Once *g*sw was obtained, water vapor flux was calculated using the product of total conductance, and vapor pressure deficit (VPD) at the leaf surface. To solve equations A1 – 3, and A22 for photosynthesis and stomatal conductance simultaneously, we used an iteration rather than a cubic equation that was used in the previous version of our model (Ueyama et al., 2016).

*A1-3. Sun/Shade radiation transfer model*

Canopy radiation transfer was based on a sun/shade model (de Pury & Farquhar, 1997; Ryu et al., 2011). The direct and diffuse portions of radiation were partitioned based on the method of Weiss & Norman (1985), for solving sun/shade radiation transfer model. Photosynthesis and transpiration were separately calculated for sun and shade leaves. An ecosystem-scale parameter of *X*eco (specifically, *Vc*max25, *J*max25, and *b*bb) can be divided into ecosystem-scale parameters of *X*sun for sun leaf and *X*shade for shade leaf.

(A23)

where *x*0 are the parameter for unit leaf area, *L*sun is sunlit leaf area index, and *L*shade is shaded leaf area index. Assuming the proximity of vertical distribution among leaf nitrogen and irradiance, *x*0 is written as:

 (A24),

 (A25),

 (A26)

where *L* is leaf area index (m2 m-2), *k*b is the extinction coefficient for beam PPFD, *k*n is the nitrogen extinction coefficient (de Pury & Farquhar, 1997; Lloyd et al., 2010), and Ω is the clumping index (Table A1; He et al., 2012). The extinction coefficient, *k*b, is calculated as

 (A27)

where *G* is the G-function defined as the projection coefficient of foliage area on the plane perpendicular to the view direction (Ross, 1981), and *θ* is the solar zenith angle. We used a look-up-table among *G* and *θ* by assuming leaf angle distribution as spherical, planophile, or erectrophile (Pisek et al., 2013).

The nitrogen extinction coefficient, *k*n, was estimated based on a relationship to maximum carboxylation rate per unit leaf area at a top of canopy, *vc*max25 (Lloyd et al., 2010).

 (A28)

By solving *k*n and *vc*max25 simultaneously, *vc*max25 was determined from big-leaf *Vc*max25 by iteratively solving Eq. A24 to A28.

*A1-4. Surface conditions at big leaf*

Surface meteorological conditions near the big-leaf canopy were estimated from micrometeorological observations. Aerodynamic conductance of sensible heat (*g*b; m s-1) was estimated as

 (A29)

where *u* is horizontal wind velocity (m s-1), *u*\* is friction velocity (m s-1), and *B*-1 is the parameter related to the roughness height (dimensionless). According to Lhomme et al. (2000), *B*-1 can be estimated using LAI (*L*; m2 m-2) as

 (A30)

where a0 (8.9667), a1 (16.127), a2 (-36.403), a3 (27.343), a4 (-9.9967), a5 (1.8013), a6 (-0.128) are empirical constants. Once *g*b was estimated, leaf temperature (*T*leaf; K) was inversely estimated using sensible heat flux (*H*; W m-2), and air temperature (*T*air; K) as

 (A31)

where cp is the specific heat (1004 J K-1 kg-1), and *ρ*air is air density (kg m-3).

Relative humidity at the leaf surface is calculated based on Su et al. (1996).

 (A32)

where *Q*air is mixing ratio (kg kg-1), *Q*leaf is mixing ratio at leaf (kg kg-1), and *g*c is canopy conductance (m s-1). *Q*leaf is calculated from saturation vapor pressure at given leaf temperature. *g*c is calculated based on the Penman-Monteith equation (Monteith, 1965).

 (A33)

where Δ is the rate of change of saturation vapor pressure with temperature (hPa K-1), γ is the psychrometric constant (hPa K-1), *D* is the vapor pressure deficits (hPa), and λE is latent heat flux (W m-2).

*A1-6. Partitioning evapotranspiration*

Measured evapotranspiration, *ET*, from the eddy covariance method was partitioned into transpiration, *T*. First, we only use dry canopy; wet conditions during rain and within one hour after rain were not used in this study. Then, dry canopy evapotranspiration was partitioned using following equations. For grassland, tundra, and all croplands except rice paddy, a known ratio between evapotranspiration and transpiration (Wang et al., 2014) was used.

 (A34)

where c1 and c2 are empirical constant. Values of c1 and c2 for grassland and tundra are 0.77 and 0.10, respectively; those for cropland are 0.91 and 0.07, respectively. For rice paddy (Sakuratani & Horie, 1985), transpiration was partitioned as:

 (A35)

For forest ecosystems, soil evaporation was estimated based on potential evaporation at the soil surface. The radiation transfer of PPFD and infrared radiation was separately calculated for estimating net radiation at the soil surface (Ryu et al., 2011), with transpiration then estimated by subtracting soil evaporation.

*A2. Optimization*

The model parameters were determined using a globally optimization method: the shuffled complex evolution method developed at the University of Arizona (SCE-UA; Duan et al., 1992, 1993, 1994). For successful global optimization, the SCE-UA introduces four concepts: a combination of random and deterministic approaches, clustering, systematic evolution, and competitive evolution. The SCE-UA was developed and used successfully to determine parameters of hydrological models. In this study, we used the method with the settings recommended in Duan et al. (1992, 1994).

The model parameters of *Vc*max25, ratio of *J*max25 to *Vc*max25 (*J*max25/*Vc*max25), *m*bb and *b*bb were determined using half-hourly GPP and λE. Parameters were determined each day using an eight-day moving window for given parameter ranges with 0.1 ~ 700 μmol m-2 s-1 for *Vc*max25, 1.8 ~ 2.7 for *J*max25/*Vc*max25 (von Caemmerer et al., 2009; Wang et al., 2007; Wullschleger, 1993), 0.1 ~ 100 for *m*bb and 0 ~ 1 mmol m-2 s-1 for *b*bb. Applying seasonally varying parameters with a moving window allows consideration of changes in physiological traits, such as periodic drought stress. First, *m*bb and *b*bb were determined by minimizing the root mean square error (RMSE) between observed and modeled transpiration, where observed GPP was used tentatively in Eq. A22 as an estimate of *A*. Coupling Eq. A22 with equations based on Fick’s law, permitted *m*bb and *b*bb to be highly constrained based on observed data. In the first optimization, ranges for *m*bb and *b*bb were determined based on standard deviations of ten determined values from randomly selected initial values. Then, *Vc*max25, *J*max25/*Vc*max25, *m*bb, and *b*bb were determined by minimizing RMSE between observed and modeled GPP, where *m*bb and *b*bb were re- determined for the range obtained from the first optimization for λE.

Ranges of parameter uncertainty were given by ten determined values from randomly selected initial values, as we found that the ten iterations for the optimization were enough for determining the global optima (Ueyama et al., 2016). We used determined parameters for subsequent analyses only when parameter uncertainty was less than 10% of the absolute value. Thus, uncertainties of the parameters associated with the optimization were less than 10% of the value.

*A3. Flux partitioning for Asian data*

We post-processed the Asian data for all sites using a standard protocol for data quality control, and flux partitioning (Ichii et al., 2017). Quality control was conducted using a spike detection method (Papale et al., 2006). Data that had been collected under calm night conditions were filtered using the friction velocity threshold (Reichstein et al., 2005). GPP was estimated as the difference between net ecosystem exchange (NEE) and ecosystem respiration (RE). Daytime RE was based on exponential relationships (Lloyd and Taylor, 1994), which were determined for each day using nighttime data with a 29-day moving window. The daily parameters of the exponential relationship were estimated using an ordinary bootstrapping procedure. In the procedure, qualified nighttime NEE were resampled 100 times without a parametric assumption as the sample size within a moving window was held. Then, RE and GPP were determined as a mean of those fluxes estimated using 100 parameters from 100 resampled data subset. This procedure was conducted using a Flux Analysis Tool program version 2 (Ueyama et al., 2012), which is open source software and is freely available from authors web-site (<http://atmenv.envi.osakafu-u.ac.jp/staff/ueyama/softwares/>).

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