

## Supporting Information

### Section 1: Comparison to stomatal optimization theories

**Katul et al (1)** give the following expression for  $f$  (their Equation 15):

$$f = 1 - \sqrt{\frac{1.6 \lambda D}{c_a}} \quad (\text{S1})$$

where in general the Katul et al. theory assumes that  $\lambda$  depends on  $C_a$ . Substituting equation S1 into our Equation (2) for  $WUE$  gives the following expression:

$$WUE = \sqrt{\frac{\lambda c_a}{1.6 D}} \quad (\text{S2})$$

In the linear approximation to the Katul et al. theory, Palmroth et al. (2) state that  $\lambda$  is directly proportional to  $C_a$ , such that:

$$WUE = k \frac{c_a}{\sqrt{D}} \quad (\text{S3})$$

where  $k$  is a constant.

**Medlyn et al. (3)** give the following expression for the  $WUE$  (their Equation 14):

$$WUE = \frac{c_a}{g_1 \sqrt{D} + D} \quad (\text{S4})$$

where  $g_1$  is a constant. In general  $g_1$  values are much larger than  $\sqrt{D}$ , so that:

$$WUE \approx \frac{c_a}{g_1 \sqrt{D}} \quad (\text{S5})$$

To derive effective  $a$  and  $b$  coefficients for each of these forms of  $WUE$  are calculated as:

$$a = \frac{\partial WUE}{\partial c_a} \frac{c_a}{WUE} \quad (\text{S6})$$

$$b = \frac{\partial WUE}{\partial D} \frac{D}{WUE} \quad (\text{S7})$$

from which we find that  $[a, b] = [1, -0.5]$  for both the models of Palmroth et al. (2) and Medlyn et al. (3).

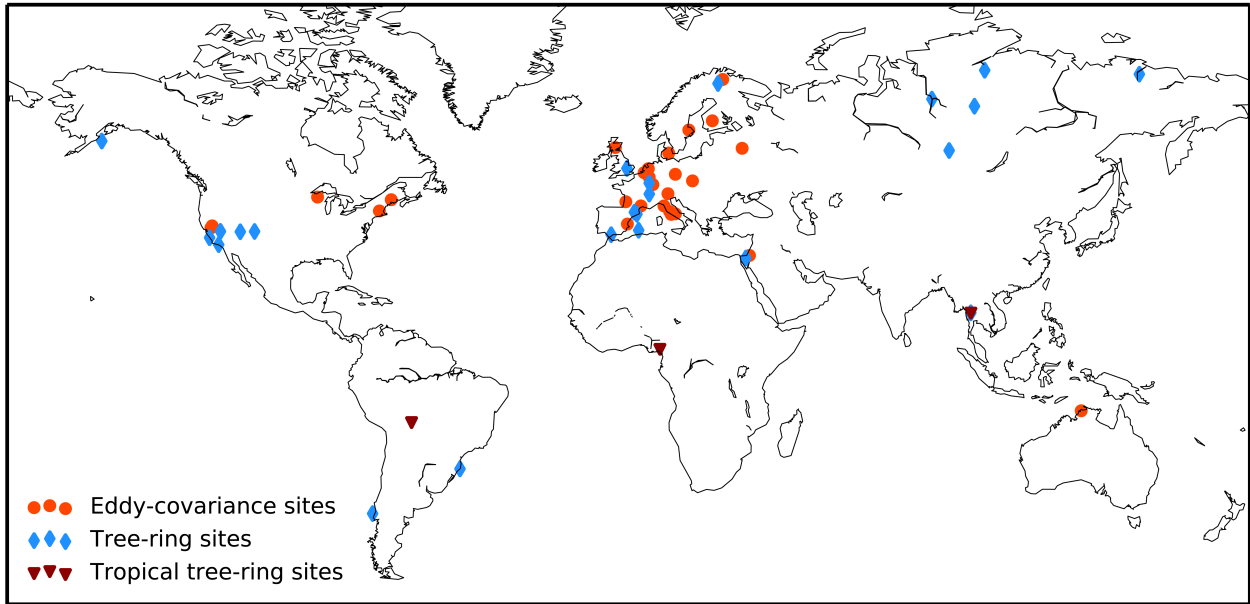
**Section 2: Partitioning the fractional change in humidity deficit a change in temperature and relative humidity**

$$D = q_{sat}(1 - RH) \tag{S8}$$

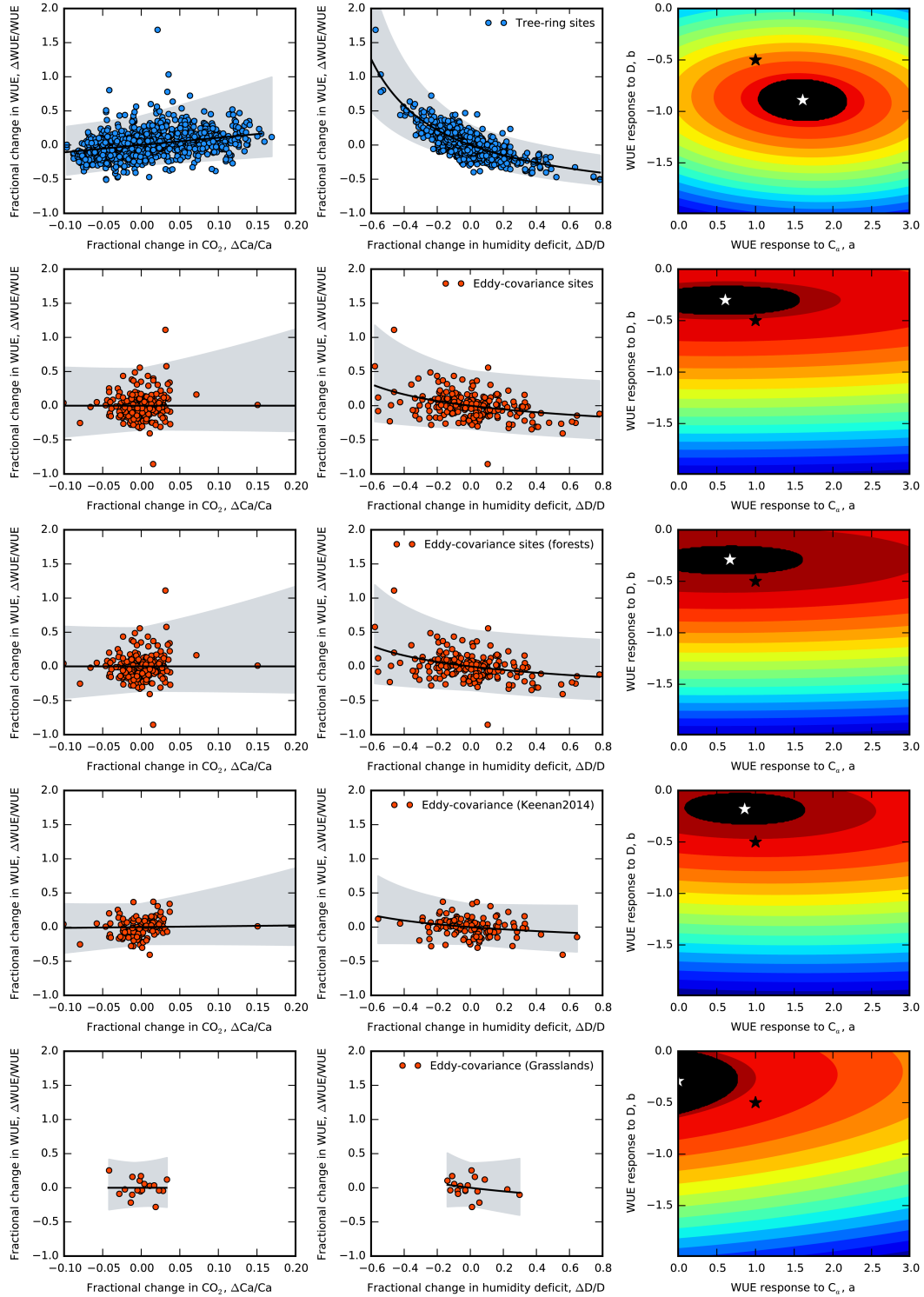
$$\Delta D = \frac{\partial q_{sat}}{\partial T}(1 - RH)\Delta T - q_{sat}\Delta RH \tag{S9}$$

$$\frac{\Delta D}{D} = \frac{1}{q_{sat}} \frac{\partial q_{sat}}{\partial T} \Delta T - \frac{\Delta RH}{(1 - RH)} \approx 0.07\Delta T - \frac{\Delta RH}{(1 - RH)} \tag{S10}$$

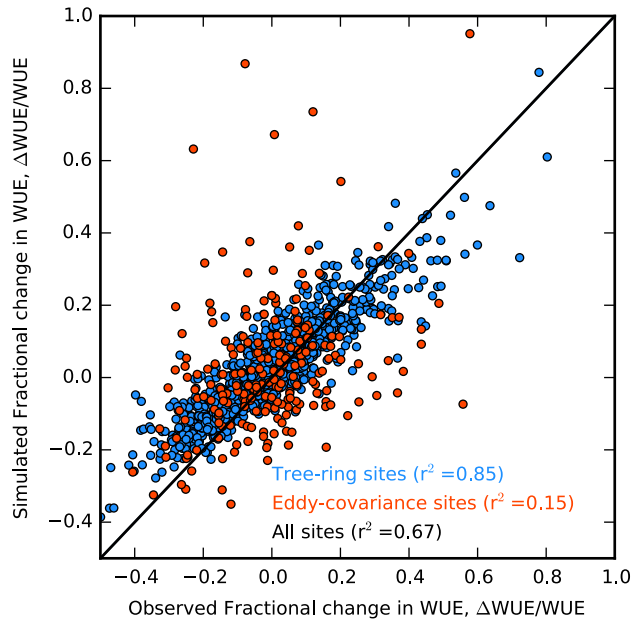
Strictly  $\Delta T$  in this equation is the surface (skin) temperature, but this is well approximated by the change in near surface air temperature, which is available from the global CRU dataset.



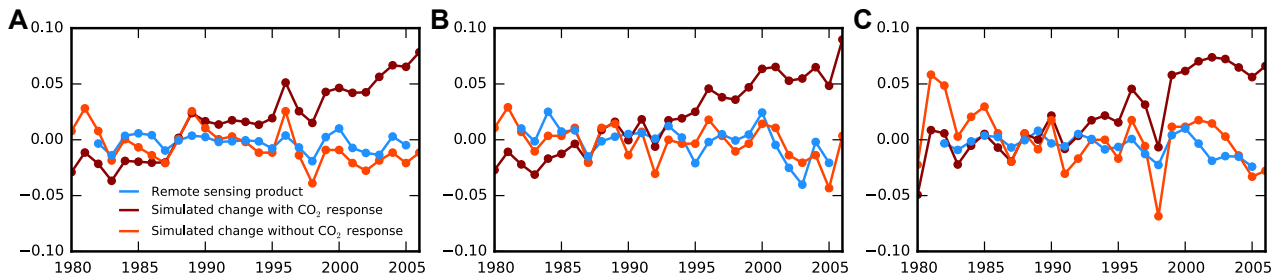
**Fig. S1.** Locations of the eddy-covariance flux sites and tree-ring sites used in Fig. 1 and 2 (see Table S2 and S3 for a list of the sites).



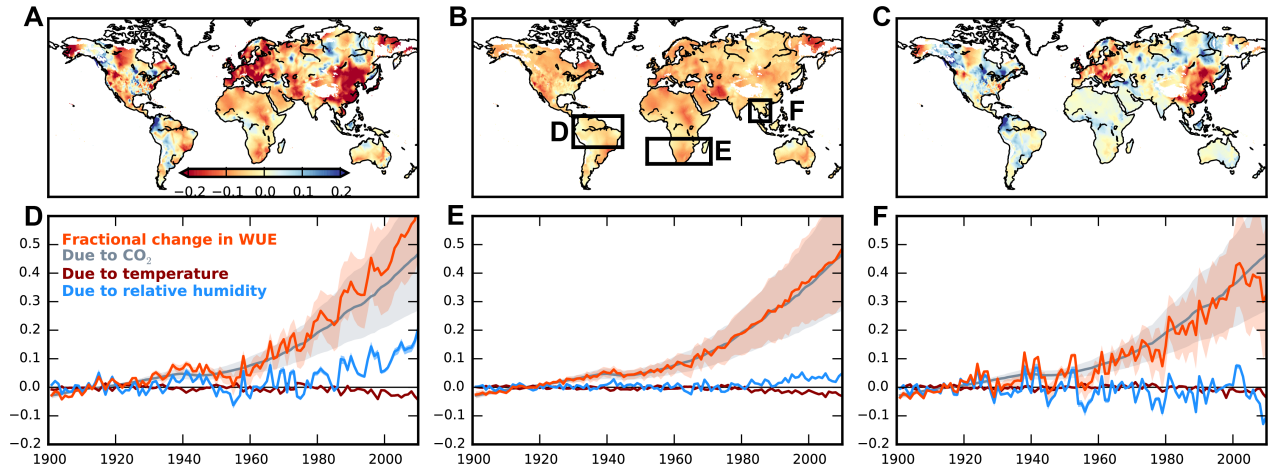
**Fig. S2.** Water Use Efficiency ( $WUE$ ) from tree-ring and eddy-covariance observations as Fig. 1, but for different groups of sites. See also Table S3 for the values of  $a$  and  $b$  for each group.



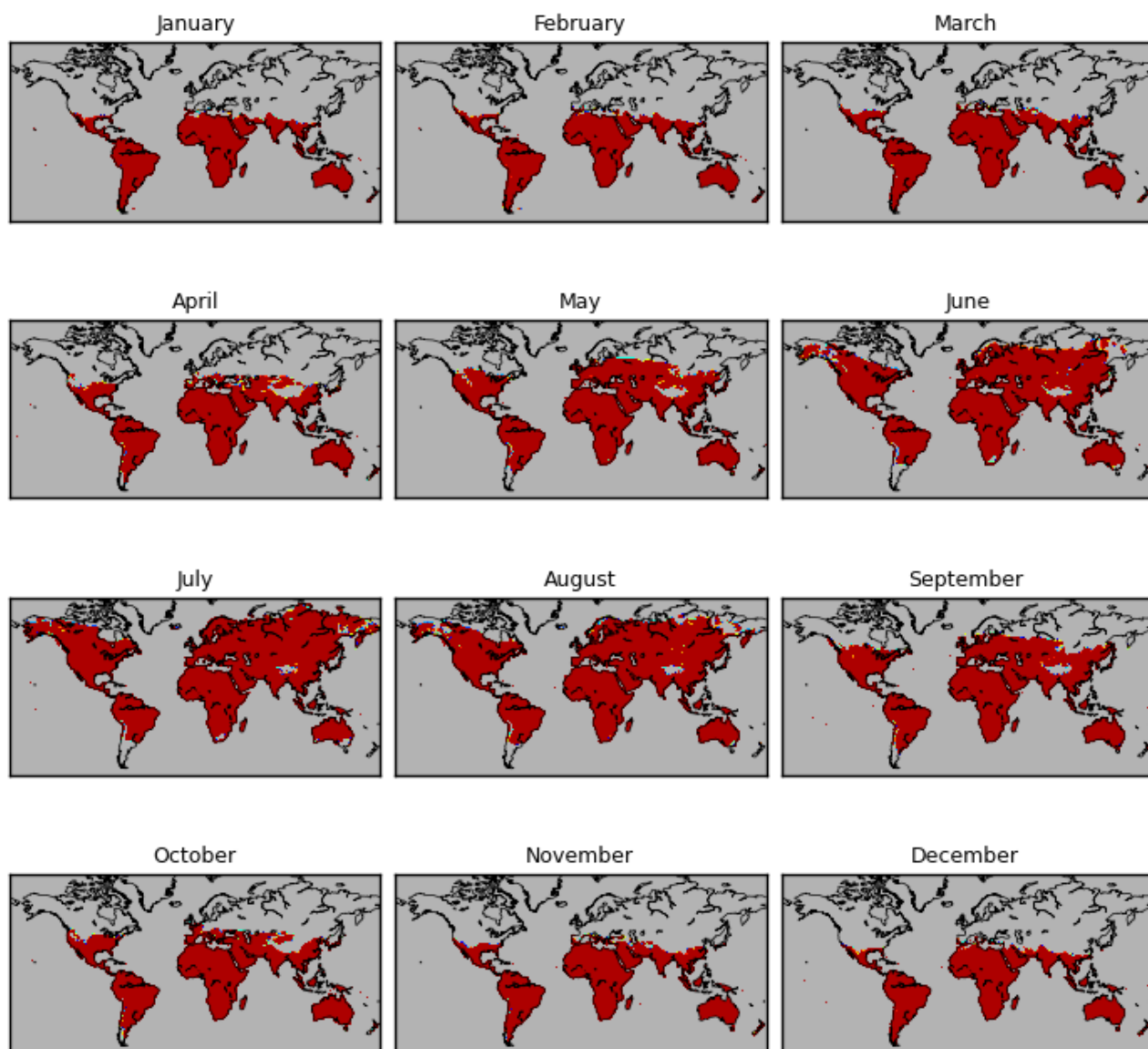
**Fig. S3.** Comparison of simulated and observed fractional change in *WUE* for the tree-ring and eddy-covariance sites.



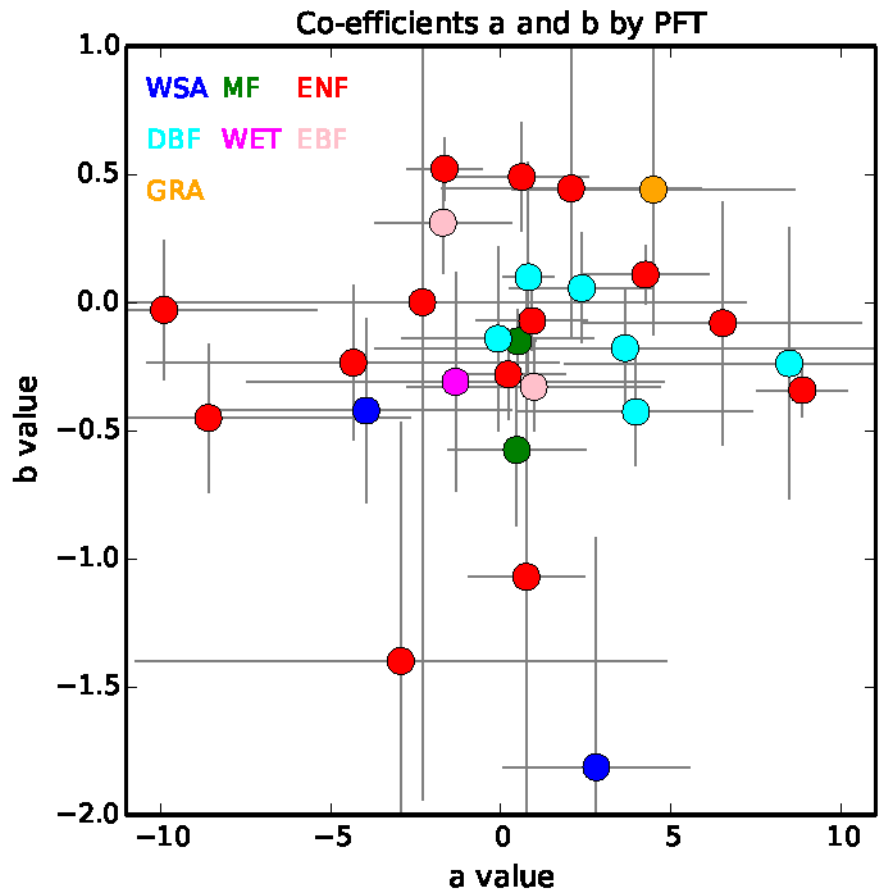
**Fig. S4.** Comparison of the estimated fractional change in *WUE* with a remote sensing product for 3 different regions: (A) the Amazon, (B) South Africa and (C) South East Asia.



**Fig. S5.** Changes in *WUE* arising from climate variables for the regions shown in Fig. S4. Spatial patterns of the fractional changes in *WUE* arising from changes in (A) climate, i.e. both temperature and relative humidity (*RH*), (B) temperature alone, and (C) *RH* alone, between 1901-1930 and 2001-2010.



**Fig S6.** Monthly mask of the area included in the growing season. For each month the average temperature (1900-1930) from the CRU TS3.20 dataset was used to determine which areas were included in the growing season based on a 10°C threshold.



**Fig S7.** Comparison of best-fit  $a$  and  $b$  parameters (see equation 3) by plant functional type (PFT). Here the sites are organized by dominant PFT, using classifications used for the FluxNet sites: evergreen broadleaf forest (EBF), evergreen needleleaf forest (ENF), grassland (GRA), mixed forest (MF), wetland (WET) and woody savannah (WSA).

**Table S1.** Summary of the Fluxnet sites selected from the *Free Fair-use* dataset (www.fluxdata.org). The sites are selected based on data availability as described in the main text.  $n$  is the number of growing seasons with data available above the threshold of 10°C,  $a$  and  $b$  are the fitted parameter values from Equation (3) and  $r^2$  is the coefficient of determination for this fit. Vegetation types are deciduous broadleaf forest (DBF), evergreen broadleaf forest (EBF), evergreen needleleaf forest (ENF), grassland (GRA), mixed forest (MF), wetland (WET) and woody savannah (WSA).

Site	Vegetation	Latitude	Longitude	$a \pm \sigma$	$b \pm \sigma$	$n$	$r^2$
AU-How	WSA	-12.49	131.15	-4.70±2.20	-2.86±1.25	6	0.63
BE-Bra	MF	51.31	4.52	-2.57±2.65	-0.76±0.23	6	0.77
BE-Vie	MF	50.31	6	0.01±0.51	-0.18±0.11	10	0.29
CZ-BK1	ENF	49.5	18.54	-10.27±6.80	-0.45±0.27	7	0.49
DE-Tha	ENF	50.96	13.57	0.68±1.79	-0.19±0.13	9	0.26
DK-Sor	DBF	55.49	11.65	0.81±0.69	0.34±0.40	10	0.20
ES-ES1	ENF	39.35	-0.32	1.24±0.74	-0.81±0.21	8	0.74
FI-Hyy	ENF	61.85	24.29	-0.25±1.47	-0.59±0.27	10	0.38
FI-Kaa	WET	69.14	27.3	4.38±5.52	-0.58±0.27	7	0.53
FR-Hes	DBF	48.67	7.06	1.69±2.56	-0.02±0.30	10	0.06
FR-LBr	ENF	44.72	-0.77	7.28±3.04	-0.10±0.25	8	0.51
FR-Pue	EBF	43.74	3.6	2.60±2.67	0.48±0.26	7	0.41
IL-Yat	ENF	31.35	35.05	-7.20±10.60	-1.00±1.29	6	0.16
IT-Col	DBF	41.85	13.59	-1.13±3.41	0.20±0.28	10	0.06
IT-Cpz	EBF	41.71	12.38	0.16±2.31	-0.48±0.13	8	0.79
IT-Ren	ENF	46.59	11.43	-10.39±4.30	0.12±0.26	8	0.65
IT-Ro1	DBF	42.41	11.93	4.00±5.59	-0.40±0.26	7	0.35
IT-SRo	ENF	43.73	10.28	7.90±1.53	-0.33±0.10	8	0.88
NL-Loo	ENF	52.17	5.74	-0.65±2.13	0.18±0.31	10	0.04
RU-Fyo	ENF	56.46	32.92	-0.45±1.89	-0.03±0.14	9	0.01
SE-Nor	ENF	60.09	17.48	-11.36±4.90	-0.84±0.30	6	0.75
UK-Gri	ENF	56.61	-3.8	-2.35±8.69	-1.10±0.94	6	0.42
US-Blo	ENF	38.9	-120.63	2.01±3.88	-1.21±0.82	10	0.29
US-Ha1	DBF	42.54	-72.17	2.81±3.07	0.46±0.26	7	0.46
US-Ho1	ENF	45.2	-68.74	3.88±1.87	0.10±0.17	9	0.38
US-Ton	WSA	38.43	-120.97	-3.42±3.58	-0.36±0.32	6	0.27
US-Var	GRA	38.41	-120.95	0.32±3.84	-0.28±0.67	6	0.05
US-WCr	DBF	45.81	-90.08	3.44±2.56	-0.44±0.21	8	0.49



**Table S2.** Summary of the tree-ring sites used in this study.  $n$  is the number of years with data available,  $a$  and  $b$  are the fitted parameter values from Equation (3) and  $r^2$  is the coefficient of determination for this fit.

Site	Species	Lat	Lon	n	$a \pm \sigma$	$b \pm \sigma$	$r^2$	Reference
Woburn Abbey - beech	<i>Fagus sylvatica</i>	52.23	-0.46	93	2.34±0.18	-0.71±0.06	0.86	Hemming et al. (1998)
Woburn Abbey - pine	<i>Pinus sylvestris</i>	52.23	-0.46	94	1.56±0.18	-0.73±0.06	0.79	Hemming et al. (1998)
Woburn Abbey - oak	<i>Quercus robur</i>	52.23	-0.46	94	1.76±0.17	-0.70±0.06	0.81	Hemming et al. (1998)
Caz	<i>Pinus nigra</i>	37.80	2.95	6	1.16±0.08	-1.08±0.09	0.98	Andreu-Hayles et al. (2011)
Urb	<i>Pinus uncinata</i>	42.23	1.70	6	0.78±0.38	-1.03±0.23	0.84	Andreu-Hayles et al. (2011)
Turuhansk	<i>Pinus sylvestris</i>	65.80	87.90	11	0.66±0.43	-1.40±0.25	0.79	Arneth et al. (2002)
Krasnoyarsk	<i>Pinus sylvestris</i>	56.00	92.90	11	1.71±0.33	-0.99±0.16	0.87	Arneth et al. (2002)
Berninger	<i>Pinus sylvestris</i>	68.46	25.93	74	0.84±0.12	-0.90±0.03	0.97	Berninger et al. (2000)
Bert select	<i>Abies alba</i>	46.52	6.02	8	2.06±0.51	-0.94±0.15	0.98	Bert et al. (1997)
Bert evenage	<i>Abies alba</i>	46.52	6.02	8	2.05±0.42	-1.02±0.13	0.98	Bert et al. (1997)
Duquesnay copp	<i>Fagus sylvatica</i>	49.00	6.00	8	0.92±0.44	-0.96±0.15	0.95	Duquesnay et al. (1998)
Duquesnay high	<i>Fagus sylvatica</i>	49.00	6.00	9	1.16±0.23	-0.90±0.09	0.97	Duquesnay et al. (1998)
Feng A	<i>Pinus edulis</i>	37.43	-112.47	15	0.40±0.15	-0.86±0.11	0.84	Feng (1999)
Feng B	<i>Pinus edulis</i>	37.50	-108.33	16	1.33±0.17	-0.91±0.17	0.83	Feng (1999)
Feng C	<i>Pinus longaeva</i>	37.53	-118.22	16	0.93±0.14	-1.04±0.11	0.91	Feng (1999)
Feng D	<i>Fitzroya cupresoides</i>	-40.05	-74.13	14	0.97±0.15	-0.92±0.05	0.98	Feng (1999)
Feng E	<i>Juniperus phoenicea</i>	30.67	34.00	13	0.91±0.18	-0.89±0.10	0.88	Feng (1999)
Feng F	<i>Pinus ponderosa</i>	35.92	-121.37	14	0.16±0.33	-0.68±0.17	0.74	Feng (1999)
Feng G	<i>Quercus lobata</i>	34.15	118.73	15	0.54±0.31	-0.97±0.10	0.89	Feng (1999)
Feng H	<i>Picea sitchensis</i>	57.92	-152.60	60	0.34±0.52	-0.94±0.06	0.98	Feng (1999)
Linares	<i>Abies pinsapo</i>	36.69	-5.02	11	1.35±0.09	-1.05±0.09	0.98	Linares et al. (2009)
Nock1	<i>Melia</i>	15.67	99.17	5	1.91±0.17	-0.45±0.24	0.99	Nock et al. (2011)
Nock2	<i>Toona</i>	15.67	99.17	10	2.22±0.18	-0.78±0.21	0.96	Nock et al. (2011)
Nock3	<i>Chukrasia</i>	15.67	99.17	10	2.02±0.19	-1.16±0.15	0.97	Nock et al. (2011)
Silva grass	<i>Araucaria angustifolia</i>	-28.59	-48.82	21	1.12±0.09	-0.99±0.12	0.93	Silva et al. (2010)
Taimyr	<i>Larix gmelinii</i>	70.63	103.20	102	1.46±0.13	-1.00±0.05	0.86	Sidorova et al. (2010)
Yakutia	<i>Larix gmelinii</i>	70.00	148.00	78	2.48±0.31	-1.01±0.09	0.74	Sidorova et al. (2010)
Siberia	<i>Larix gmelinii</i>	64.53	100.23	102	1.82±0.05	-1.00±0.03	0.97	Sidorova et al. (2009)
Penuelas central	<i>Fagus sylvatica</i>	41.77	2.43	29	2.29±0.19	-1.01±0.10	0.91	Penuelas et al. (2008)
Penuelas high	<i>Fagus sylvatica</i>	41.77	2.43	29	1.77±0.18	-1.07±0.10	0.89	Penuelas et al. (2008)
Penuelas low	<i>Fagus sylvatica</i>	41.77	2.43	25	0.94±0.15	-0.97±0.08	0.90	Penuelas et al. (2008)

**Table S3.** Comparison of  $a$  and  $b$  values from Equations 3 and 4 between tree-ring and eddy-covariance sites from Tables S2 and S3. The values of the combined datasets are compared to the values of the tree-ring or eddy-covariance sites only. The range is derived from the black areas in Fig. S2.

<b>Description</b>	<b><math>a</math></b>	<b><math>b</math></b>
Tree-rings + Eddy-covariance	$1.51 \pm 0.57$	$-0.72 \pm 0.16$
Tree-rings	$1.61 \pm 0.54$	$-0.89 \pm 0.17$
Eddy-covariance	$0.79 \pm 0.79$	$-0.30 \pm 0.10$
Eddy-covariance (Keenan)	$1.46 \pm 0.78$	$-0.19 \pm 0.10$
Eddy-covariance (Forests)	$0.90 \pm 0.90$	$-0.29 \pm 0.10$
Eddy-covariance (Grasslands)	$0.00 \pm 0.00$	$-0.29 \pm 0.29$

**Table S4.** Comparison of the quality of the fits to observed  $WUE$  changes at *Fluxnet* sites, using varying numbers of environmental predictor variables. Our default model (equation 6) which uses just atmospheric  $CO_2$  concentration ( $C_a$ ) and atmospheric humidity deficit ( $D$ ), is given by the first row of the table. The additional environmental predictors used are those readily available from a significant number ( $n$ ) of *Fluxnet* sites: solar radiation at the surface ( $R_g$ ), air temperature ( $T_a$ ), and soil water content ( $SWC$ ). The adjusted  $r^2$  value accounts for the increase in the basic  $r^2$  measure that is to be expected when additional parameters are introduced into a statistical model. Models with larger adjusted- $r^2$  values are therefore considered more robust, as they avoid over-fitting of the data.

<b>Predictor Variables</b>	<b><math>n</math></b>	<b><math>a</math></b>	<b><math>b</math></b>	<b><math>r^2</math></b>	<b>Adjusted <math>r^2</math></b>
$C_a, D$	227	1.028	-0.290	0.103	0.096
$C_a, D, R_g$	227	1.056	-0.309	0.105	0.093
$C_a, D, R_g, T_a$	227	1.124	-0.331	0.108	0.092
$C_a, D, R_g, T_a, SWC$	124	0.532	-0.416	0.118	0.081

**Table S5.** Fractional change in WUE simulated in the historical simulations of CMIP5 Earth System Models (ESMs). These are for research centres and models where both transpiration (“tran”) and Gross Primary Productivity (“gpp”) monthly variables are available, with ensemble size given in the 3<sup>rd</sup> column. The year 2005 normalised change (4<sup>th</sup> column) is relative to the mean over the modelled period from 1860 to 1890.

<b>Research Centre</b>	<b>Climate Model</b>	<b>Ensemble size</b>	<b>Fractional change in WUE by 2005</b>
BCC	bcc-csm1-1	3	0.19
BCC	bcc-csm1-1-m	1	0.21
BNU	BNU-ESM	1	0.20
CCCma	CanESM2	5	0.10
CMCC	CMCC-CESM	1	0.02
INM	inmcm4	1	0.07
IPSL	IPSL-CM5A-LR	6	0.15
IPSL	IPSL-CM5A-MR	3	0.14
IPSL	IPSL-CM5B-LR	1	0.16
MIROC	MIROC-ESM	3	0.03
MIROC	MIROC-ESM-CHEM	1	0.03
MPI-M	MPI-ESM-LR	3	0.24
MPI-M	MPI-ESM-MR	3	0.21
MPI-M	MPI-ESM-P	2	0.21
MRI	MRI-ESM1	1	0.28
NASA-GISS	GISS-E2-H	8	0.13
NASA-GISS	GISS-E2-H-CC	1	0.16
NASA-GISS	GISS-E2-R	20	0.13
NASA-GISS	GISS-E2-R-CC	1	0.11
NCAR	CCSM4	8	0.12
NCC	NorESM1-M	3	0.10
NCC	NorESM1-ME	1	0.10
NOAA-GFDL	GFDL-ESM2G	1	0.20
NOAA-GFDL	GFDL-ESM2M	1	0.19
NSF-DOE-NCAR	CESM1-BGC	1	0.12
NSF-DOE-NCAR	CESM1-CAM5	3	0.17
NSF-DOE-NCAR	CESM1-FASTCHEM	3	0.12
NSF-DOE-NCAR	CESM1-WACCM	1	0.13
<b>Ensemble Mean</b>			<b>0.14</b>

### References in Supplementary Information

- 1 Katul G, Manzoni S, Palmroth S, Oren R (2010) A stomatal optimization theory to describe the effects of atmospheric CO<sub>2</sub> on leaf photosynthesis and transpiration. *Annals of Botany* 105:431–442.
- 2 Palmroth S et al. (2013) On the complementary relationship between marginal nitrogen and water-use efficiencies among *Pinus taeda* leaves grown under ambient and CO<sub>2</sub>-enriched environments. *Annals of Botany* 111:467–477.
- 3 Medlyn BE et al. (2011) Reconciling the optimal and empirical approaches to modelling stomatal conductance. *Global Change Biol* 17:2134–2144.