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Supplement of

Large but decreasing effect of ozone on the European carbon sink

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Supplementary Information

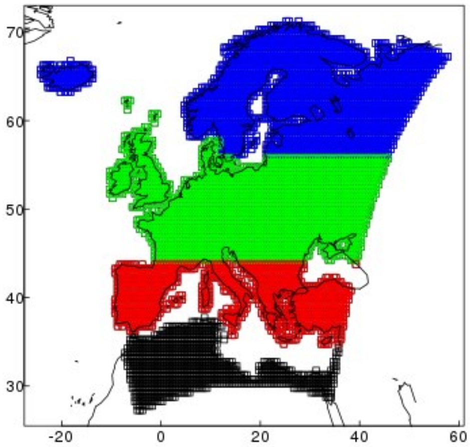

Figure S1. Regions of model domain, blue is Boreal, green is Temperate, red is Mediterranean.

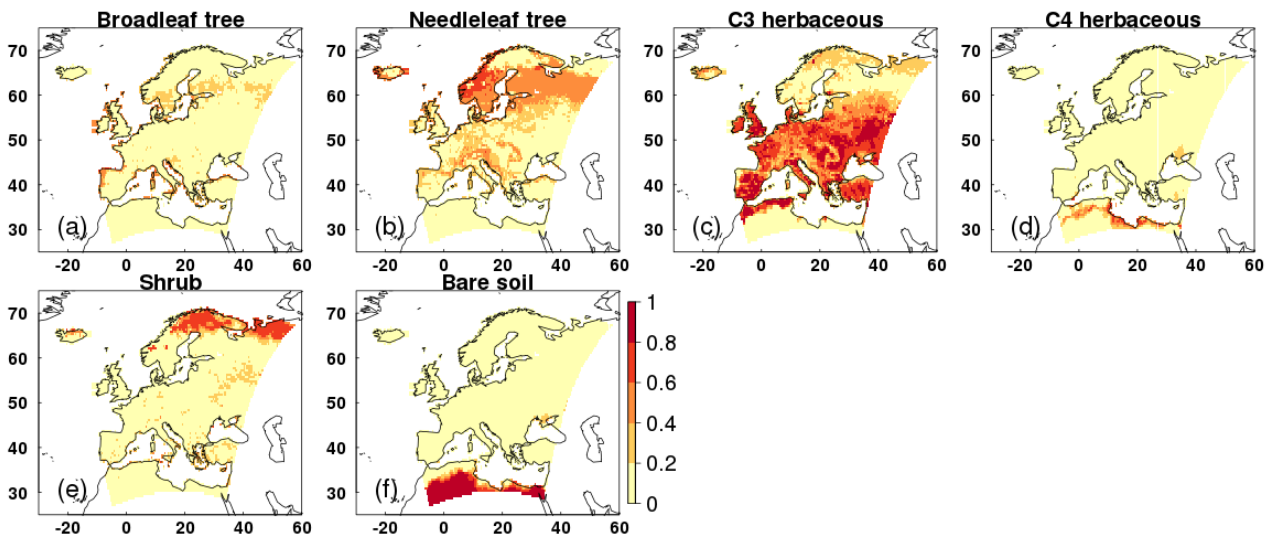

Figure S2. Fractional cover of each JULES PFT and bare soil at 0.5º x 0.5º resolution.

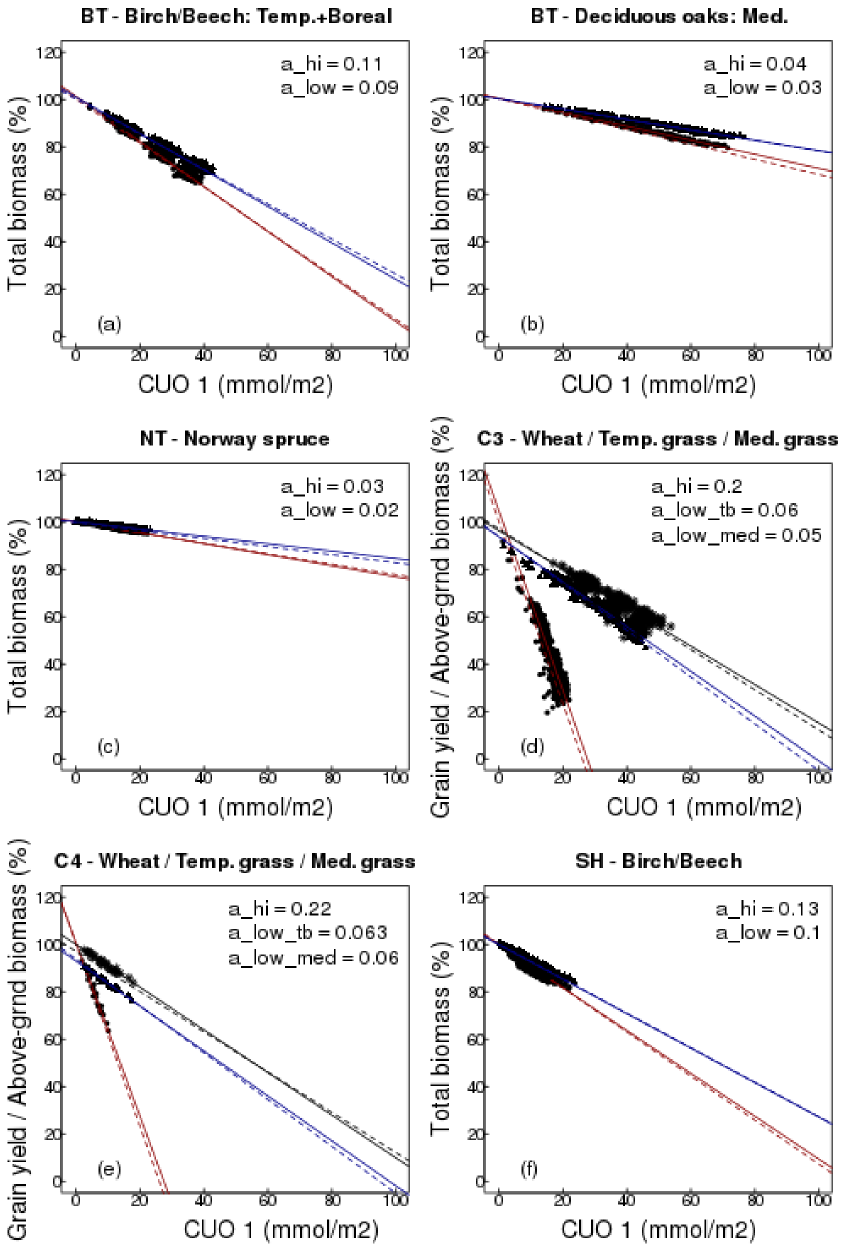

Figure S3. Calibration of JULES for O_3 impacts on plant productivity for each JULES PFT ; a) broadleaf tree – temperate/boreal, b) broadleaf tree Mediterranean, c) Needle leaf tree, d) C_3 herbaceous (split into temperate/boreal and Mediterranean for the natural grasslands), e) C_4 herbaceous (split into temperate/boreal and Mediterranean for the natural grasslands), and f) shrub. High (red) and low (blue) plant O_3 sensitivities are shown. For the herbaceous PFTs the low sensitivity calibration is separate for Mediterranean regions (black). The solid line is the regression line through the modelled points, the dashed line is the regression line from the observed dose-response relationship. The x axis is cumulative uptake of O_3 (CUO) above the critical O_3 threshold (F_{O3crit}).

	High Sensitivity				
	BT	NT	C3	C4	SH
F_{O3crit} (nmol/m²/s)	1.00	1.00	1.00	1.00	1.00
a (mmol/m²)	0.110	0.030	0.200	0.220	0.130
Function	Birch/Beech: y=100.2-0.93x	Norway spruce: y=99.8-0.22x	Wheat: y=100.3-3.85x	Wheat: y=100.3-3.85x	Birch/Beech: y=100.2-0.93x
dqcrit (kg kg⁻¹)	0.09	0.06	0.1	0.075	0.1
f0	0.875	0.875	0.9	0.8	0.9
g_1 *(kPa$^{0.5}$)*	3.22	2.22	5.56	1.1	2.24
	Low Sensitivity				
	BT	**NT**	**C3**	**C4**	**SH**
F_{O3crit} (nmol/m²/s)	1.00	1.00	1.00	1.00	1.00
a (mmol/m²)	0.090	0.020	0.060	0.063	0.100
Function	Birch/Beech: y=100.2-0.74x	Norway spruce: y=99.8-0.17x	Temperate perennial grassland: y=93.9-0.99x	Temperate perennial grassland: y=93.9-0.99x	Birch/Beech: y=100.2-0.74x
	High Sensitivity				
	BT - Med.				
F_{O3crit} (nmol/m²/s)	1.00				
a (mmol/m²)	0.040				
Function	Dec. Oaks: y=100.3-0.32x				
	Low Sensitivity				
	BT - Med.	**C3 - Med.**	**C4 - Med.**		
F_{O3crit} (nmol/m²/s)	1.00	1.00	1.00		
a (mmol/m²)	0.030	0.050	0.060		
Function	Dec. Oaks: y=100.3-0.22x	Mediterranean annual pasture: y=97.1-0.85x	Mediterranean annual pasture: y=97.1-0.85x		

Table S1. PFT-specific parameter values used in the O_3 uptake and g_s formulation in JULES. F_{O3crit} is the critical O_3 threshold above which damage occurs, *a* determines the reduction in photosynthesis with O_3 exposure, 'function' shows the regression equation for the observed functions (x is F_{O3crit}), dq_{crit} (kg kg^{-1}) is a PFT specific parameters representing the critical humidity deficit at the leaf surface (used in the default JULES g_s model), f_0 is the leaf internal to atmospheric CO_2 ratio (c_i/c_a) at the leaf specific humidity deficit (also used in the default JULES g_s model), and g_1 is the PFT specific parameter of the Medlyn *et al*., (2011) g_s model. The parameters dq_{crit}, f_0 and g_1 vary by PFT, but not by O_3 sensitivity so are only shown once here.

Site name	Country	Latitude	Longitude	Simulated years	Land cover	Dominant PFT(s)
US-UMB	USA	45.56	-84.71	2000-2014	Broadleaf forest	100% BT
IT-CA1	Italy	42.38	12.02	2011-2014	Broadleaf forest	100% BT
FI-Hyy	Finland	62	24.3	1996-2014	Needleleaf forest	100% NT
DE-Tha	Germany	51	13.57	1996-2014	Needleleaf forest	100% NT
CH_Cha	Switzerland	47.21	8.41	2006-2014	C3 grassland	80% C3, 20% bare soil
US-SRG	USA	31.8	-110.83	2008-2014	C4 grassland	80% C4, 20% bare soil
CG-Tch	Congo	-4.5	11.66	2006-2009	Deciduous savanna	50% BT, 15% C4, 25% shrub, 10% bare soil

Table S2. Sites from the FLUXNET2015 dataset used in the site simulations to evaluate *gs* models.

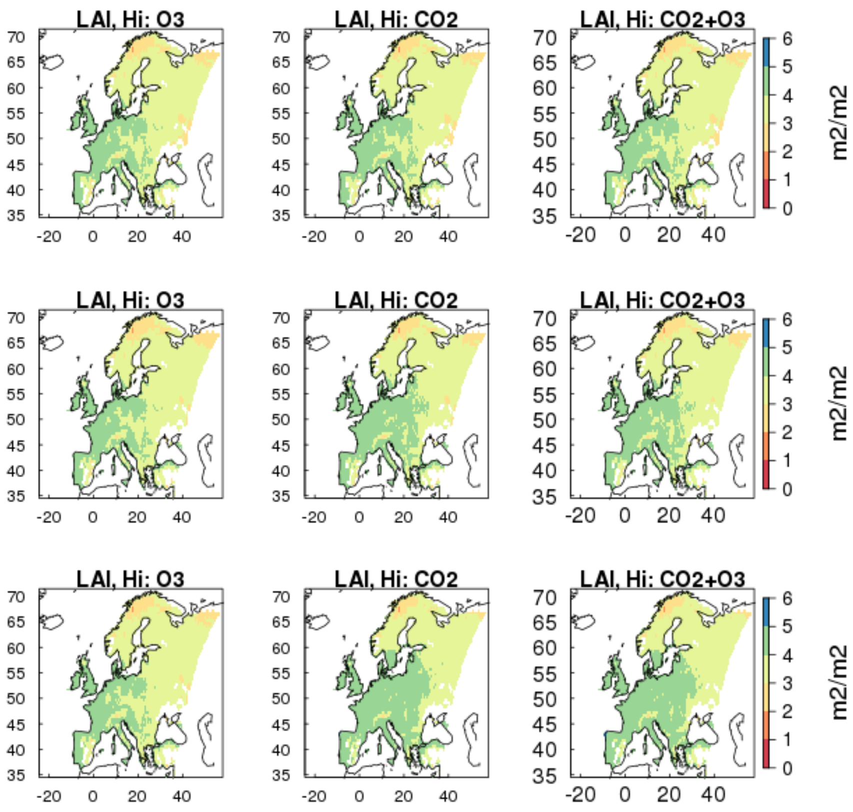

Figure. S4. Simulated mean annual leaf area index (LAI) in1901 (top row), 2001 (middle row) and 2050 (bottom row) for the high plant O_3 sensitivity.

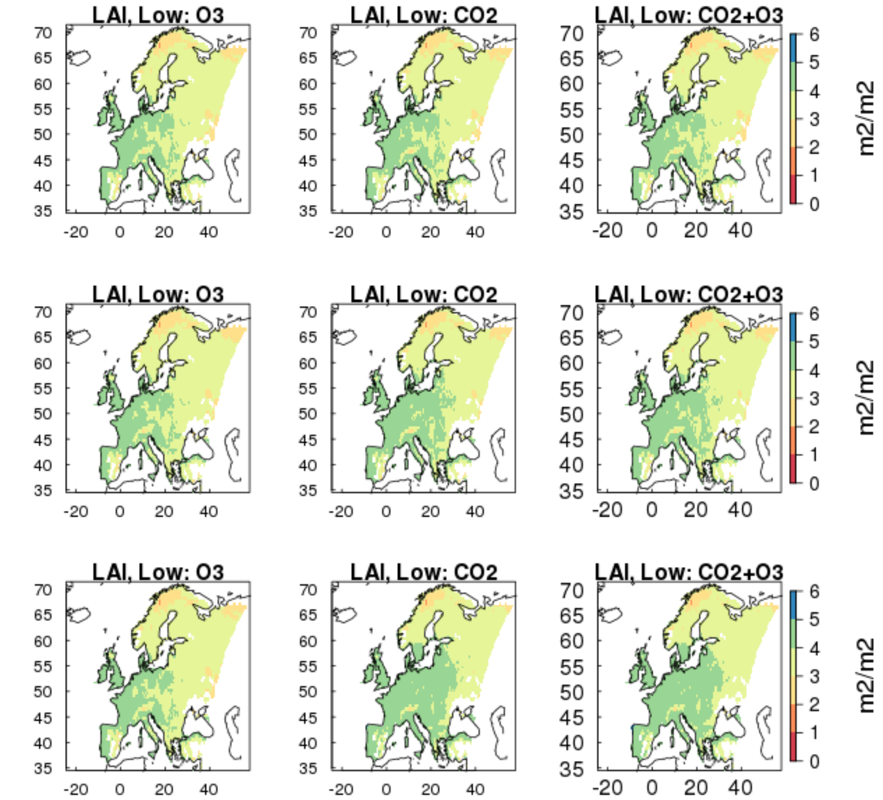

Figure. S5. Simulated mean annual leaf area index (LAI) in1901 (top row), 2001 (middle row) and 2050 (bottom row) for the low plant O_3 sensitivity.

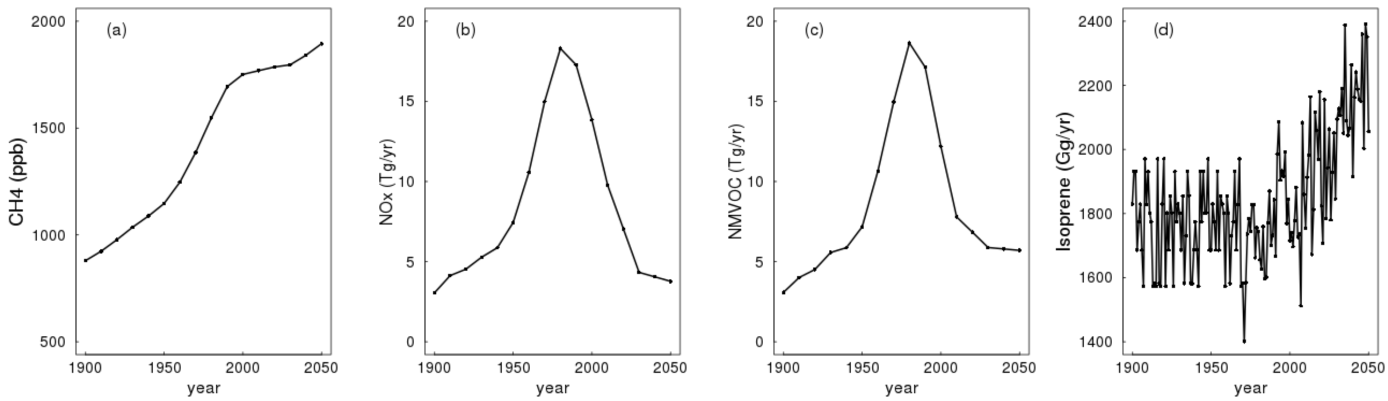

Figure. S6. Trend in the emissions of the O_3 precursors a) methane (CH_4 (ppb)), b) nitrous oxides (NO_x (Tg/yr)), c) non-methane VOC (NMVOC (Tg/yr)) and d) Isoprene (Gg/yr) over Europe from 1900 to 2050.

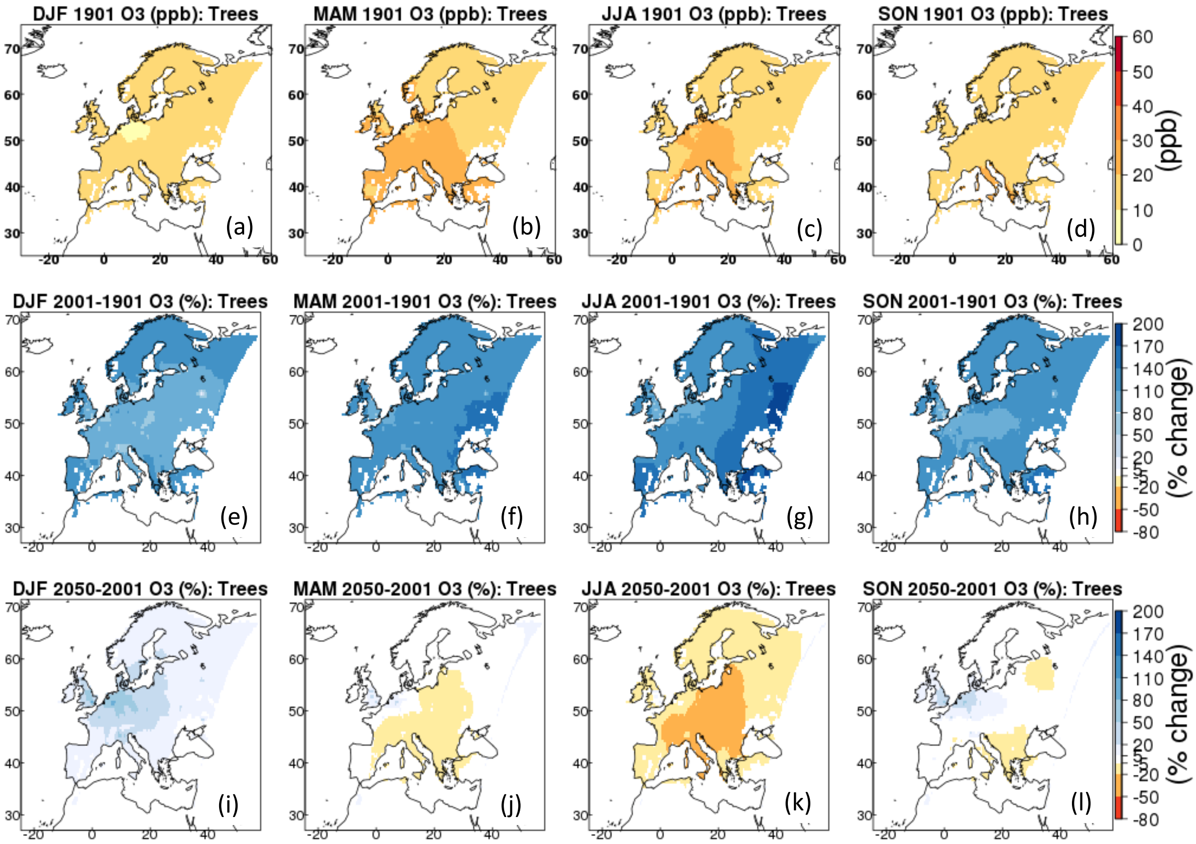

Figure S7. (a-d) 1901 seasonal mean (DJF, MAM, JJA, SON) O_3 concentration (ppb) from EMEP for woody (tree and shrub) PFTs; (e-h) change in seasonal O_3 concentration (%) from 1901 to 2001; (i-l) change in seasonal O_3 concentration (%) from 2001 to 2050.

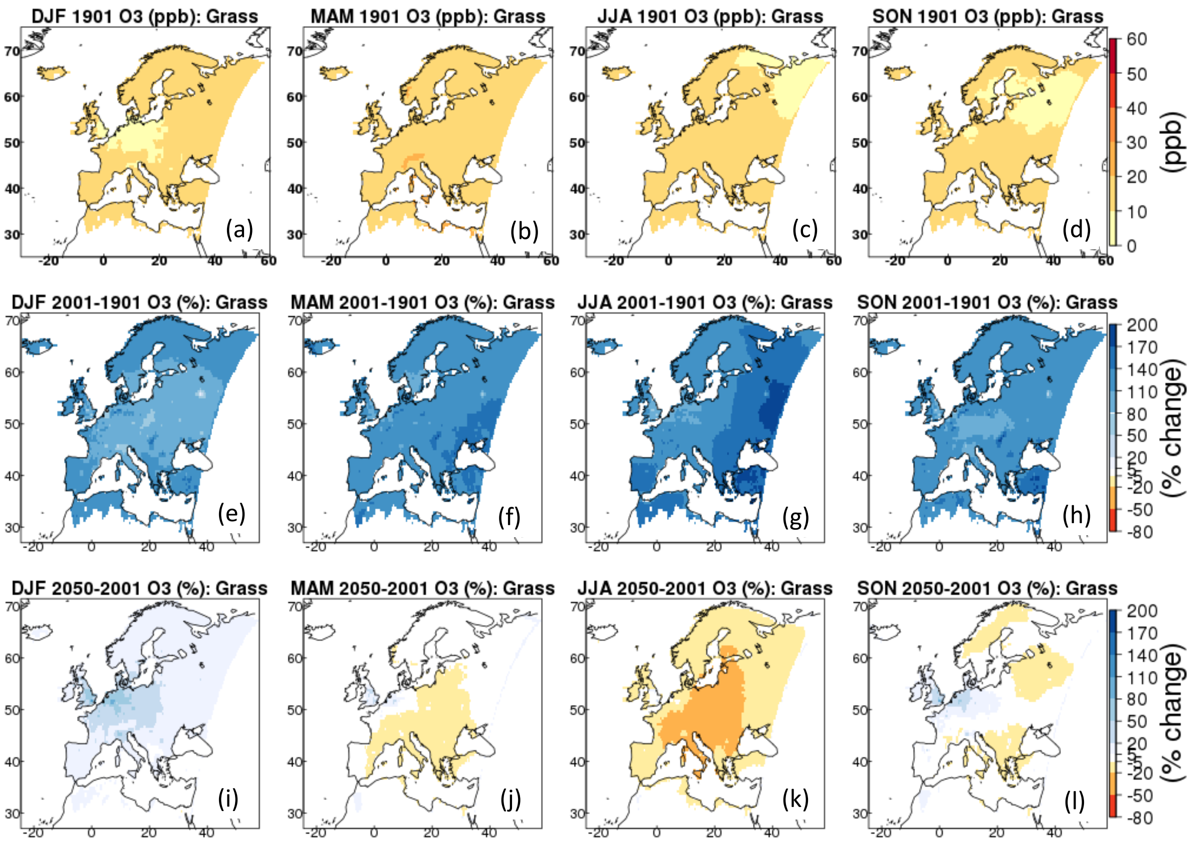

Figure S8. (a-d) 1901 seasonal mean (DJF, MAM, JJA, SON) O_3 concentration (ppb) from EMEP for herbaceous PFTs; (e-h) change in seasonal O_3 concentration (%) from 1901 to 2001; (i-l) change in seasonal O_3 concentration (%) from 2001 to 2050.

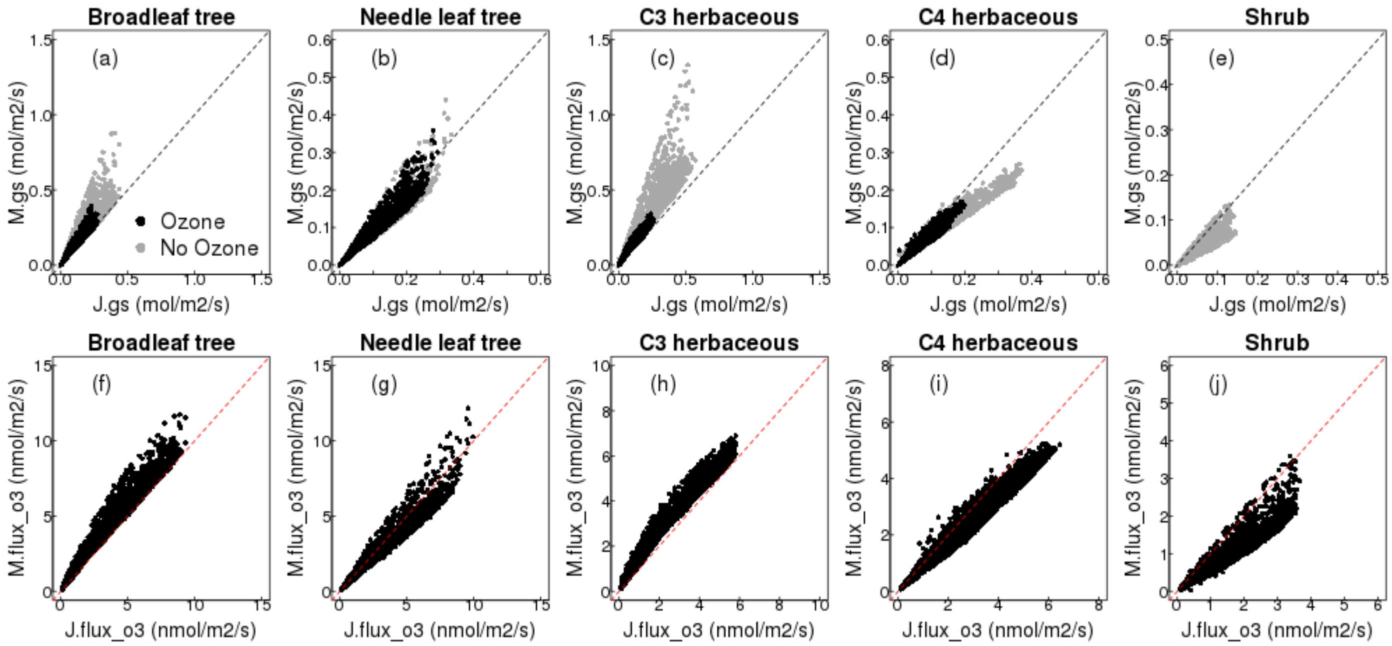

Figure S9. Comparison of the Medlyn *et al*., (2011) g_s model (y axis) versus the Jacobs (1994) g_s model (x axis) currently used in JULES for all five JULES PFTs, for stomatal conductance (*gs*, top row) and the flux of O_3 through the stomata (flux_o3, bottom row) for a dry site (high soil moisture stress).

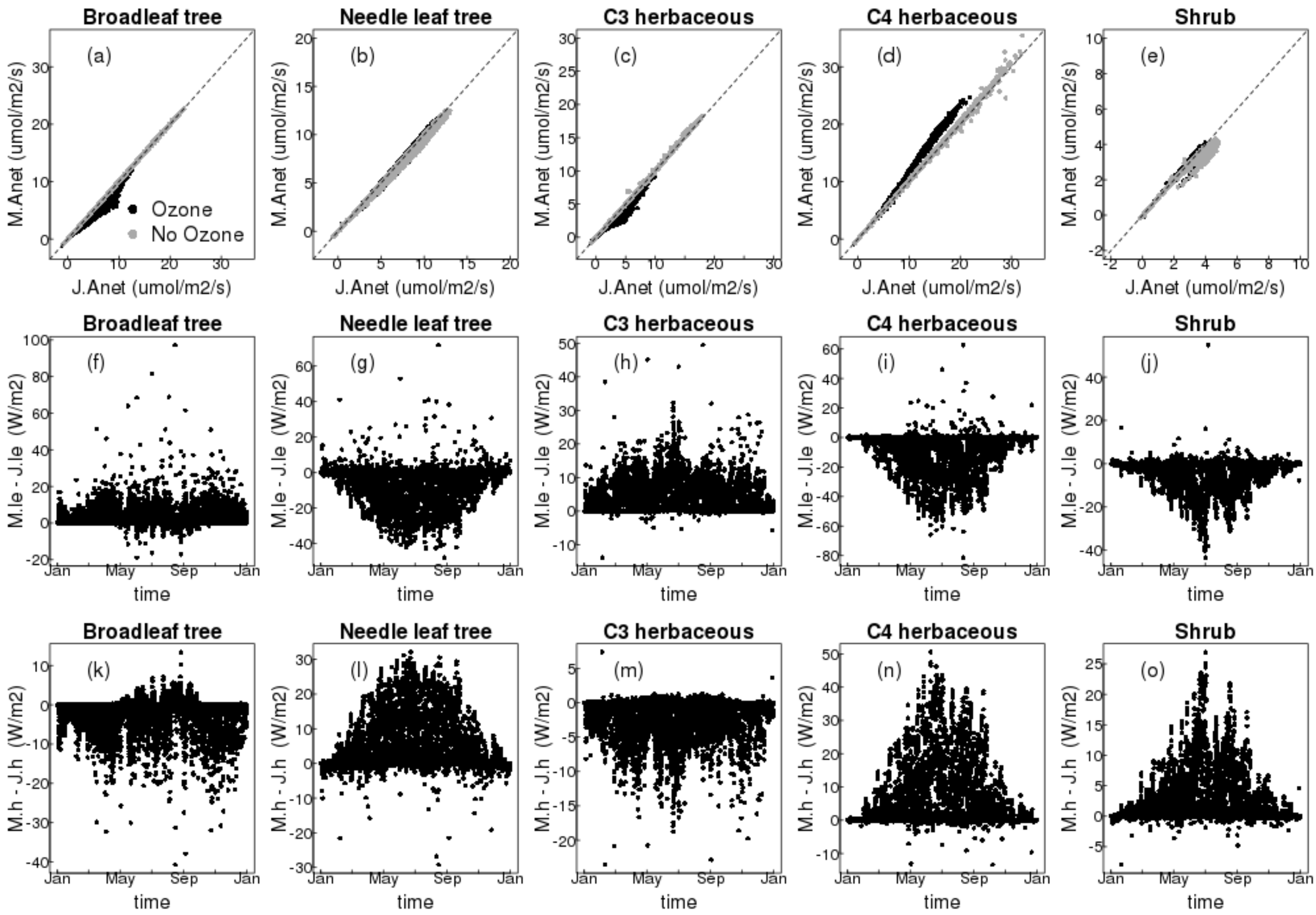

Figure S10. Comparison of the Medlyn *et al*., (2011) g_s model (y axis) versus the Jacobs (1994) g_s model (x axis) currently used in JULES for all five JULES PFTs at the wet site (low soil moisture stress), for net photosynthesis (*Anet*, top row). Residual plots (Medlyn - Jacobs) show the difference between models over the year for latent heat (le, middle row) and sensible heat (h, bottom row).

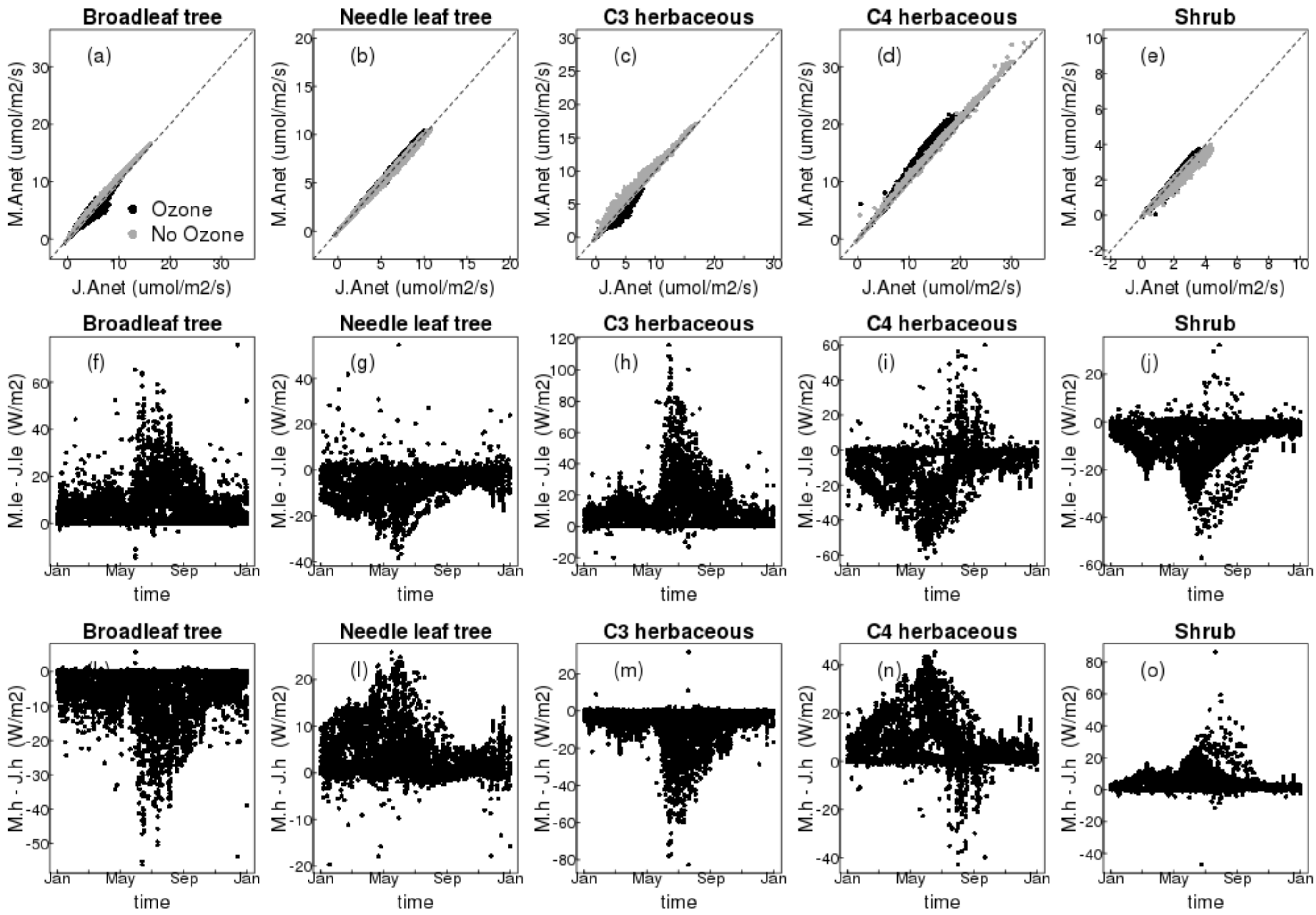

Figure S11. Comparison of the Medlyn *et al*., (2011) g_s model (y axis) versus the Jacobs (1994) g_s model (x axis) currently used in JULES for all five JULES PFTs at the dry site (high soil moisture stress), for net photosynthesis (*Anet*, top row). Residual plots (Medlyn - Jacobs) show the difference between models over the year for latent heat (le, middle row) and sensible heat (h, bottom row).

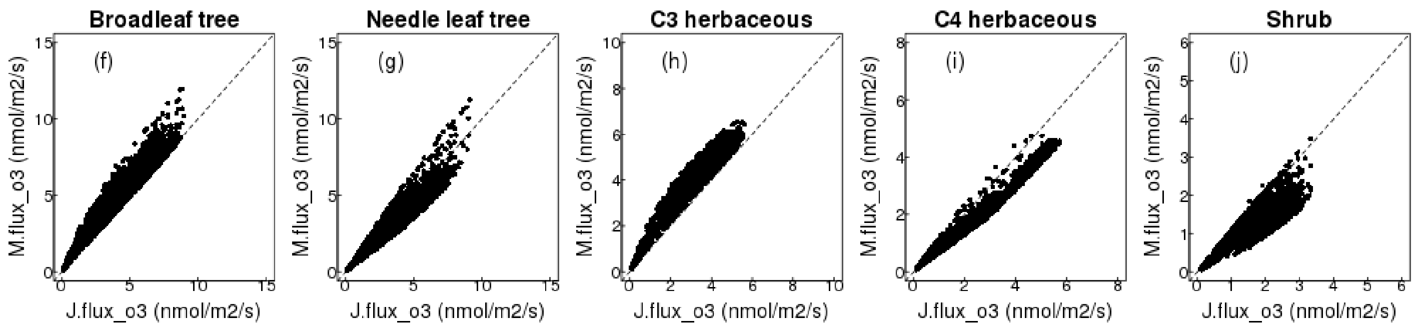

Figure S12. Comparison of the flux of O_3 through the stomata (flux_o3) for the Medlyn *et al*., (2011) g_s model (y axis) versus the Jacobs (1994) g_s model (x axis) currently used in JULES for all five JULES PFTs for the wet site (low soil moisture stress).

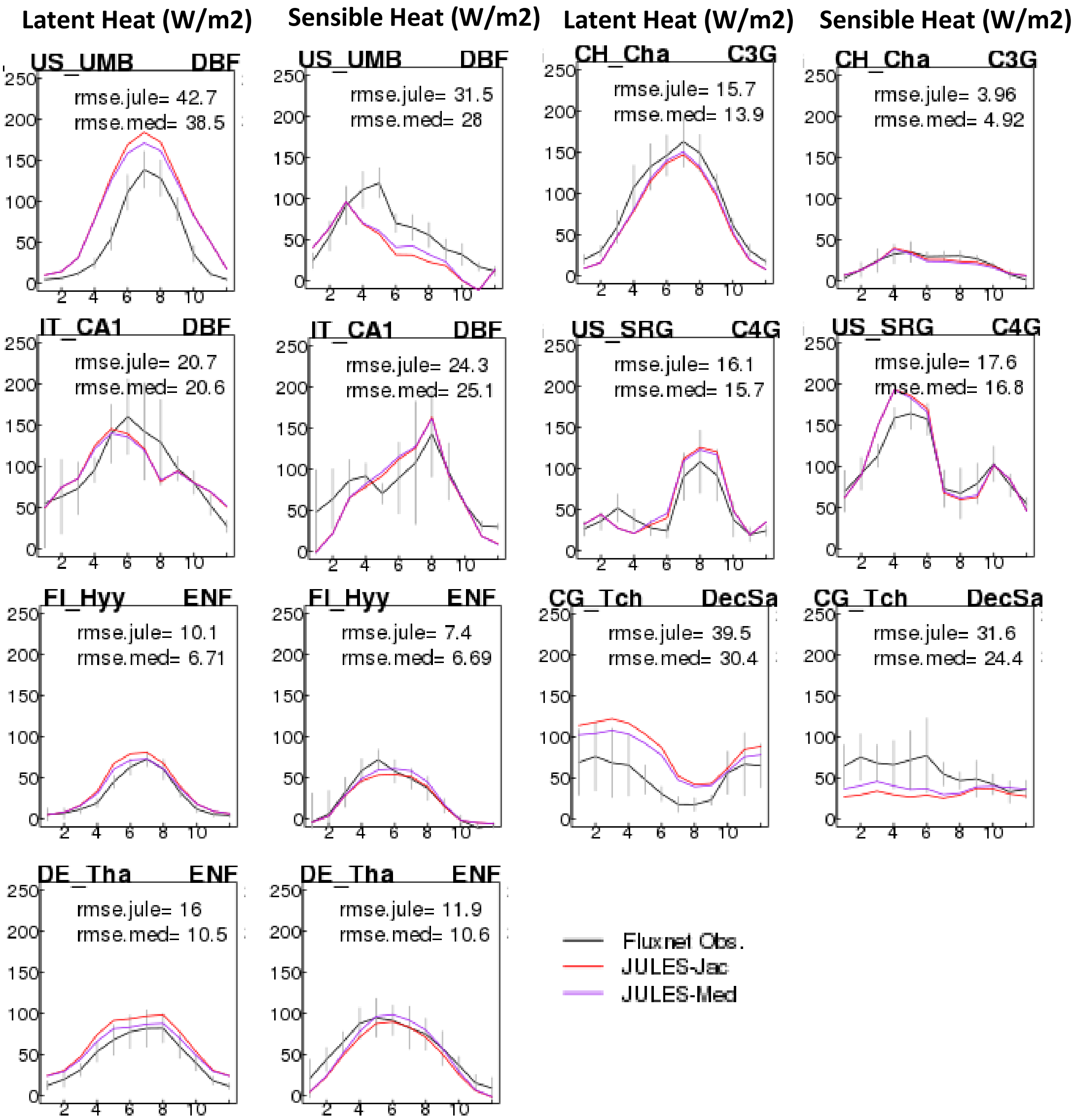

Fig. S13 Monthly mean fluxes of latent and sensible heat. Observations ± standard deviation from FLUXNET2015 sites are shown as black line with grey vertical bars, JULES with the JAC g_s model is shown in red and JULES with the MED g_s model are shown in purple. Also shown are the root mean squared error (rmse) for each simulation.

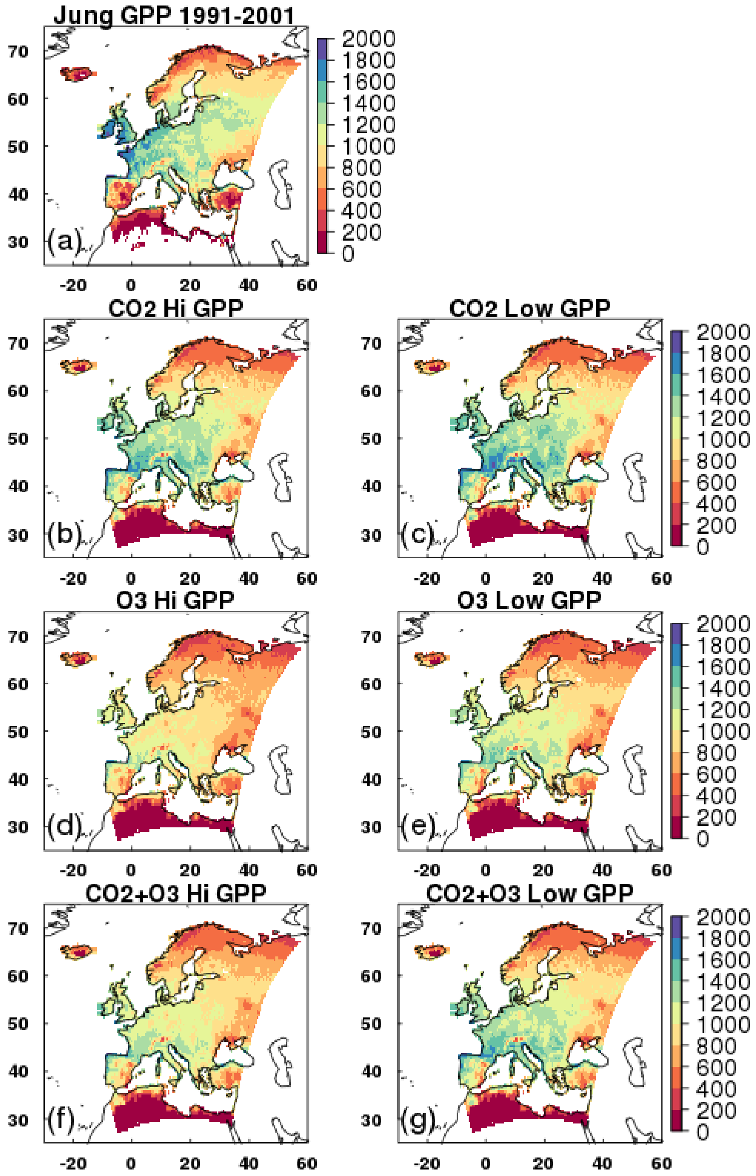

Figure S14. Mean GPP (g C m^2 yr^{-1}) from 1991 to 2001 for a) the observations based globally extrapolated Flux Network model tree ensemble (MTE) (Jung et al., 2011); b, c) model simulations with transient CO_2 and fixed O_3; d, e)model simulations with fixed CO_2 and transient O_3, and f, g) our model simulations with transient CO_2 and transient O_3. All model simulations show GPP for high and low plant O_3 sensitivity respectively.

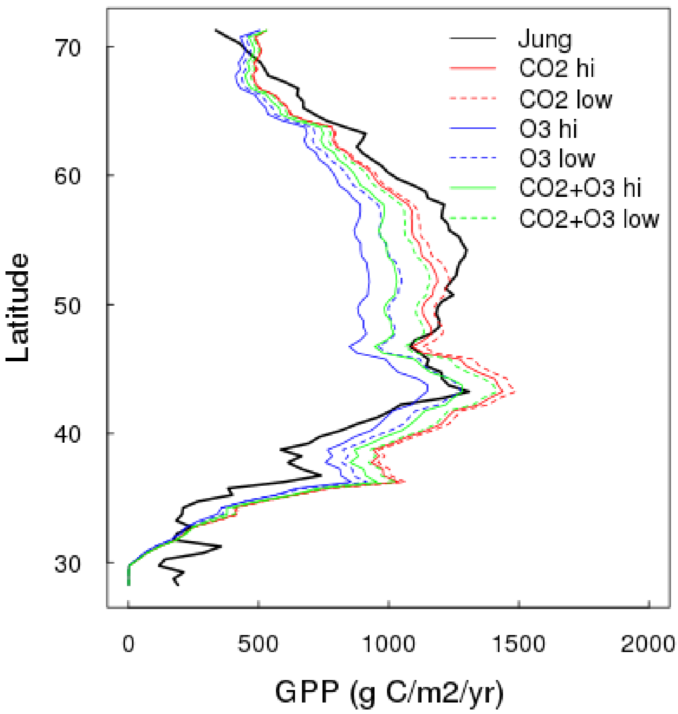

Figure S15. Zonal mean GPP from 1991 to 2001 for FLUXNET-MTE (Jung) and all JULES scenario simulations with both high (solid lines) and low (dashed lines) plant O_3 sensitivity.

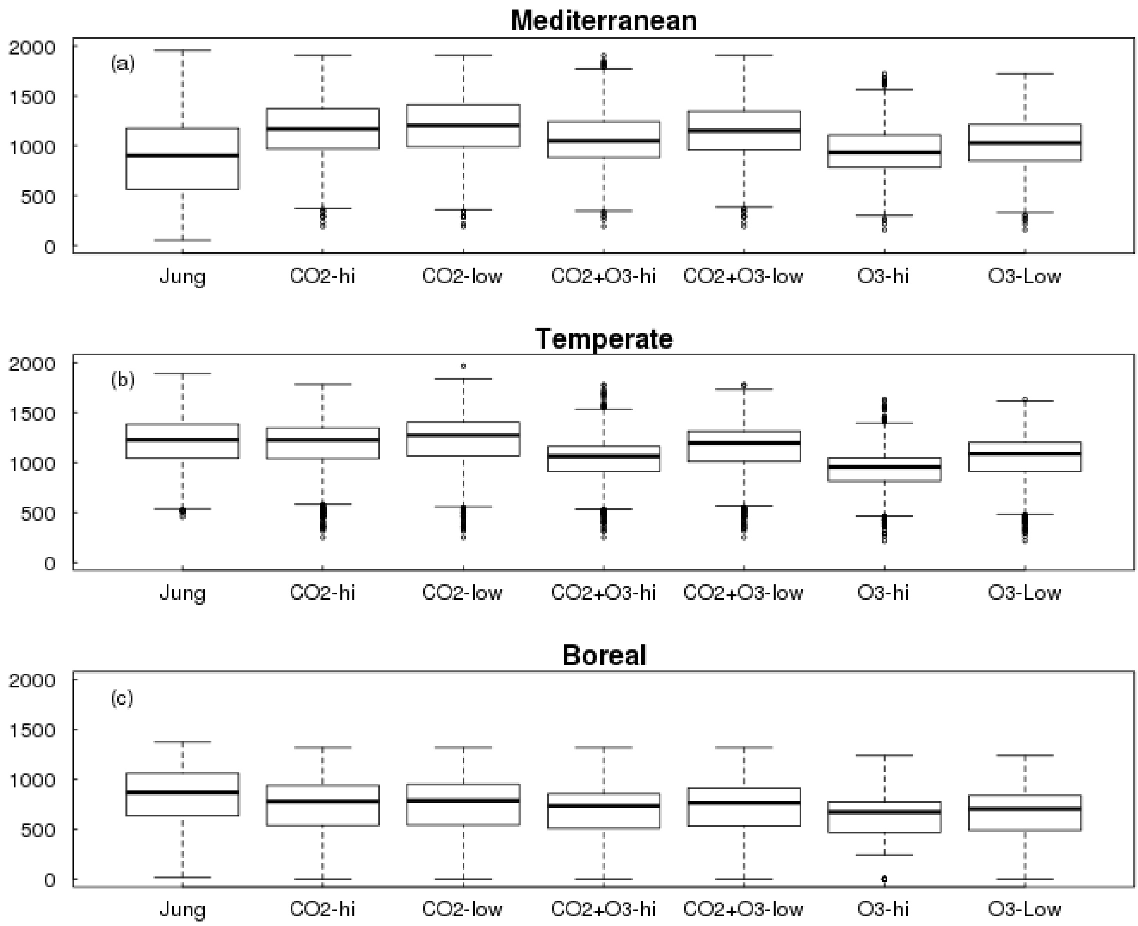

Figure S16. Mean GPP from 1991 to 2001 shown by region, comparing MTE (Jung) and all JULES scenario simulations with both high and low plant O_3 sensitivity.

	Future run, constant climate (1901 - 2001)					
	Hi Sensitivity					
	GPP (Pg C yr^{-1})	NPP (Pg C yr^{-1})	g_s (m/s)	Veg C (Pg C)	Soil C (Pg C)	Land C (Pg C)
Value in 1901:	9.05	4.46	0.03228	41.1	125.8	167
Absolute diff. (2001 - 1901):						
O_3	-0.81	-0.47	0.00	-0.02	-9.09	-9.21
CO_2	1.16	0.76	0.00	2.82	1.52	4.24
CO_2 + O_3	0.13	0.12	0.00	2.37	-5.55	-3.28
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	-8.95	-10.54	-8.55	-0.05	-7.23	-5.51
CO_2	12.82	17.04	-6.07	6.86	1.21	2.54
CO_2 + O_3	1.44	2.69	-13.66	5.77	-4.41	-1.96
	Lower Sensitivity					
	GPP (Pg C yr^{-1})	NPP (Pg C yr^{-1})	g_s (m/s)	Veg C (Pg C)	Soil C (Pg C)	Land C (Pg C)
Value in 1901:	9.34	4.65	0.03319	41.1	126.4	167.5
Absolute diff. (2001 - 1901):						
O_3	-0.30	-0.21	0.00	-0.21	-3.38	-3.59
CO_2	1.15	0.74	0.00	2.73	3.70	6.43
CO_2 + O_3	0.65	0.43	0.00	2.21	0.29	2.50
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	-3.21	-4.52	-3.31	-0.51	-2.67	-2.14
CO_2	12.31	15.91	-6.39	6.64	2.93	3.84
CO_2 + O_3	6.96	9.25	-9.88	5.38	0.23	1.49

Table S3. Simulated changes in the European land carbon cycle due to changing O_3 and CO_2 concentrations. Shown are changes in total carbon stocks (Land C), split into vegetation (Veg C) and soil (Soil C) carbon, and gross primary productivity (GPP), net primary productivity (NPP) and conductance (g_s), between 1901 and 2001

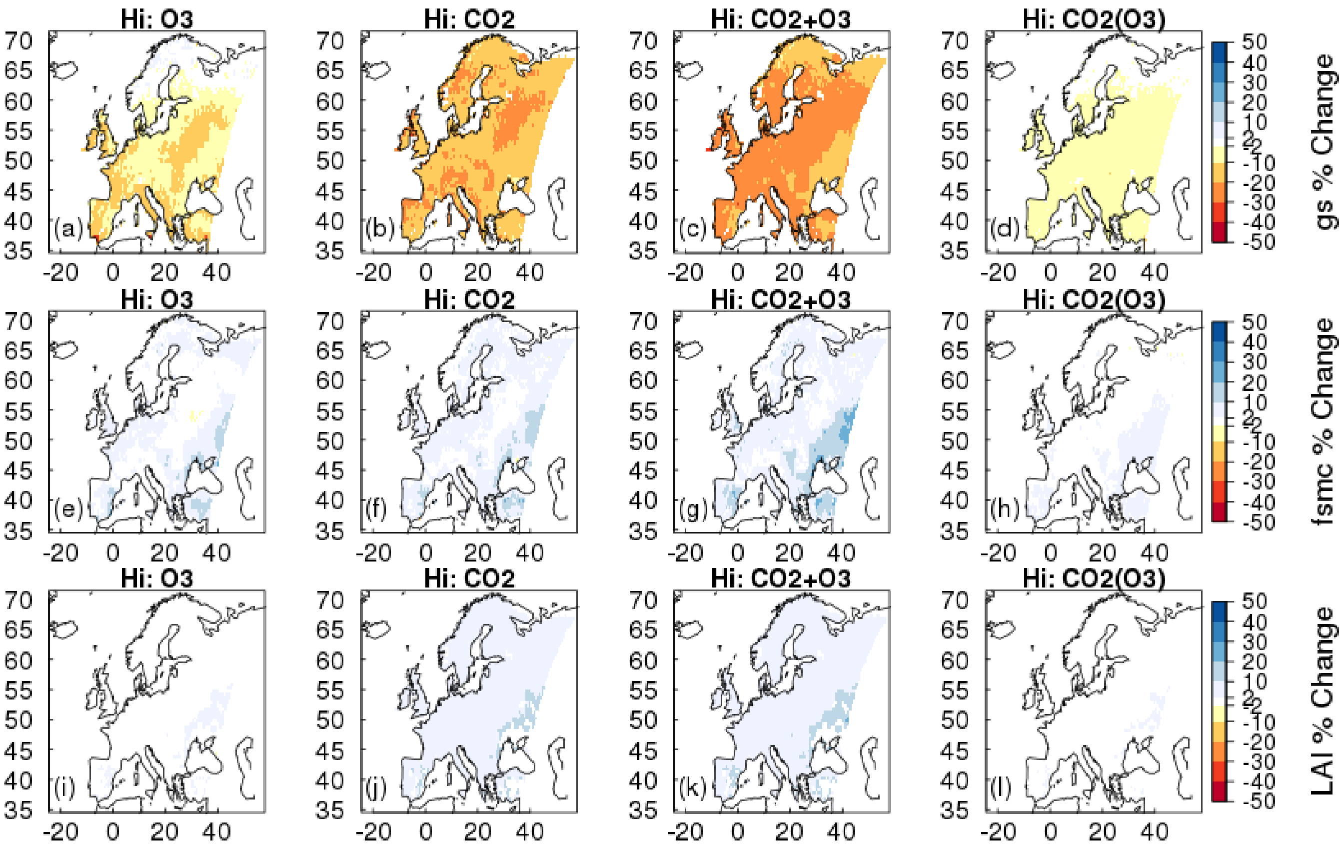

Figure S17. Simulated percentage change in stomatal conductance (gs) a-c), soil moisture availability factor (fsmc) d-e) and leaf area index (*LAI*) g-i) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3), CO_2 effects at fixed pre-industrial O_3 concentration (CO2), and effects of CO_2 and O_3 changing simultaneously (CO2+O3). Changes are shown for the periods 1901 to 2050 for the higher plant O_3 sensitivity.

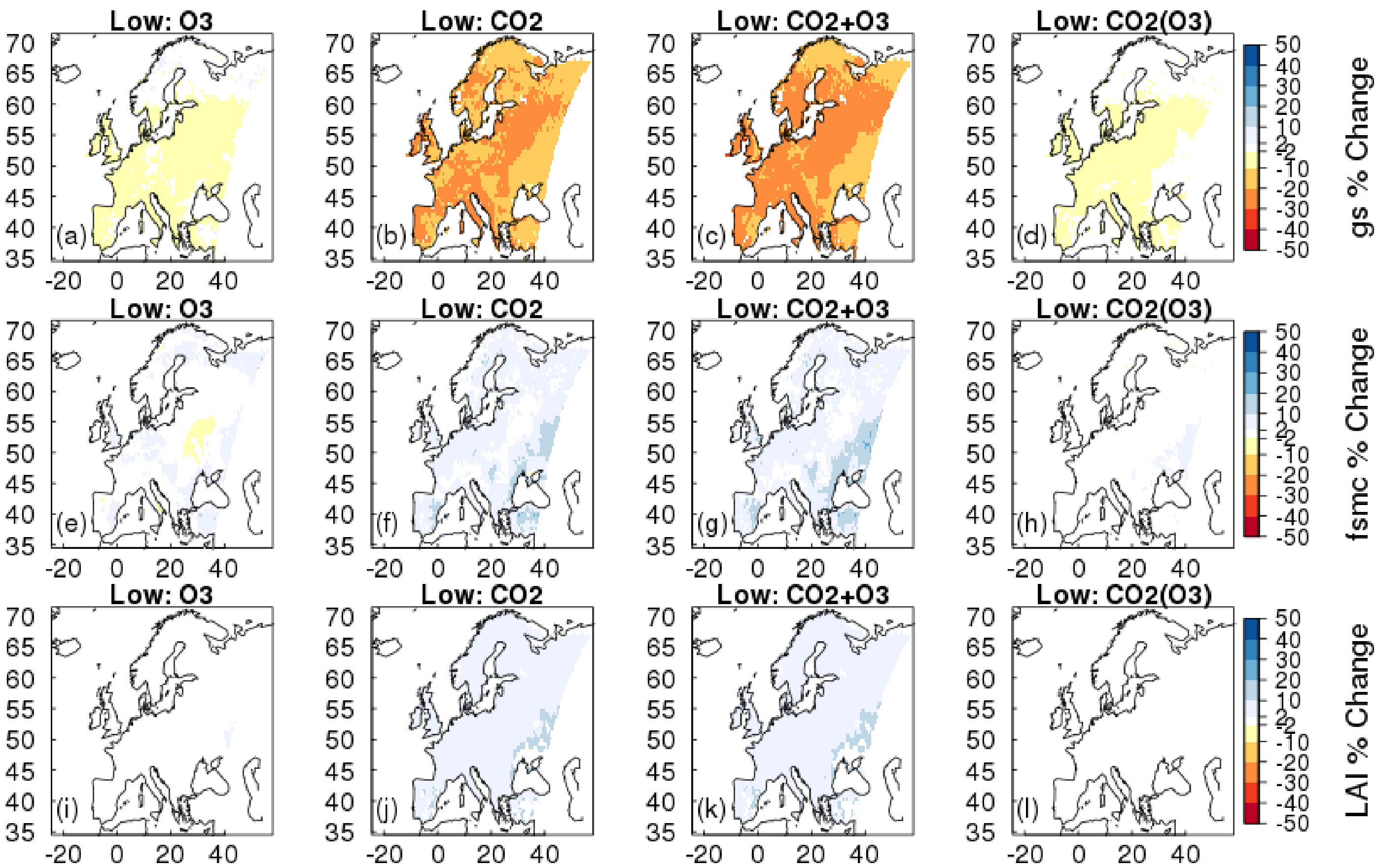

Figure S18. Simulated percentage change in stomatal conductance (gs) a-c), soil moisture availability factor (fsmc) d-e) and leaf area index (*LAI*) g-i) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O3), CO_2 effects at fixed pre-industrial O_3 concentration (CO2), and effects of CO_2 and O_3 changing simultaneously (CO2+O3). Changes are shown for the periods 1901 to 2050 for the lower plant O_3 sensitivity.

	GPP	NPP	g_s	Veg C	Soil C	Land C
Future run, constant climate (2001 - 2050)						
Hi Sensitivity						
	(Pg C yr^{-1})	(Pg C yr^{-1})	(m/s)	(Pg C)	(Pg C)	(Pg C)
Value in 2001:						
O_3	8.24	3.99	0.02952	41.08	116.71	157.79
CO_2	10.21	5.22	0.03032	43.92	127.32	171.24
CO_2 + O_3	9.18	4.58	0.02787	43.47	120.25	163.72
Absolute diff. (2050 - 2001):						
O_3	0.01	0.00	0.00	-0.09	-2.35	-2.44
CO_2	1.42	0.95	0.00	5.25	7.73	12.98
CO_2 + O_3	1.66	1.07	0.00	5.11	6.00	11.11
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	0.12	0.00	0.00	-0.22	-2.01	-1.55
CO_2	13.91	18.20	-13.89	11.95	6.07	7.58
CO_2 + O_3	18.08	23.36	-11.37	11.76	4.99	6.79
Lower Sensitivity						
	GPP	NPP	g_s	Veg C	Soil C	Land C
	(Pg C yr^{-1})	(Pg C yr^{-1})	(m/s)	(Pg C)	(Pg C)	(Pg C)
Value in 2001:						
O_3	9.04	4.44	0.03	40.89	123.02	163.91
CO_2	10.49	5.39	0.03	43.83	130.1	173.93
CO_2 + O_3	9.99	5.08	0.02991	43.31	126.69	170
Absolute diff. (2050 - 2001):						
O_3	0.02	-0.06	0.00	-0.13	-0.94	-1.07
CO_2	1.35	0.92	0.00	5.25	7.89	13.14
CO_2 + O_3	1.50	1.00	0.00	5.11	7.25	12.35
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	0.22	-1.35	-0.72	-0.32	-0.76	-0.65
CO_2	12.87	17.07	-14.64	11.98	6.06	7.55
CO_2 + O_3	15.02	19.69	-13.37	11.80	5.72	7.26

Table S4. Simulated changes in the European land carbon cycle due to changing O_3 and CO_2 concentrations. Shown are changes in total carbon stocks (Land C), split into vegetation (Veg C) and soil (Soil C) carbon, and gross primary productivity (GPP), net primary productivity (NPP) and conductance (g_s), between 2001 and 2050.

Future run, constant climate (1901 - 2050)

	Hi Sensitivity					
	GPP (Pg C yr^{-1})	NPP (Pg C yr^{-1})	g_s (m/s)	Veg C (Pg C)	Soil C (Pg C)	Land C (Pg C)
Value in 1901:	9.05	4.46	0.03228	41.1	125.8	167
Absolute diff. (2050 - 1901):						
O_3	-0.80	-0.47	0.00	-0.11	-11.44	-11.65
CO_2	2.58	1.71	-0.01	8.07	9.25	17.22
CO_2 + O_3	1.79	1.19	-0.01	7.48	0.45	7.83
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	-8.84	-10.54	-8.55	-0.27	-9.09	-6.98
CO_2	28.51	38.34	-19.11	19.64	7.35	10.31
CO_2 + O_3	19.78	26.68	-23.48	18.20	0.36	4.69

	Lower Sensitivity					
	GPP (Pg C yr^{-1})	NPP (Pg C yr^{-1})	g_s (m/s)	Veg C (Pg C)	Soil C (Pg C)	Land C (Pg C)
Value in 1901:	9.34	4.65	0.03319	41.1	126.4	167.5
Absolute diff. (2050 - 1901):						
O_3	-0.40	-0.27	0.00	-0.34	-4.32	-4.66
CO_2	2.50	1.66	-0.01	7.98	11.59	19.57
CO_2 + O_3	2.15	1.43	-0.01	7.32	7.54	14.85
Relative diff. (%)	(%)	(%)	(%)	(%)	(%)	(%)
O_3	-4.28	-5.81	-4.01	-0.83	-3.42	-2.78
CO_2	26.77	35.70	-20.10	19.42	9.17	11.68
CO_2 + O_3	23.02	30.75	-21.93	17.81	5.97	8.87

Table S5. Simulated changes in the European land carbon cycle due to changing O_3 and CO_2 concentrations. Shown are changes in total carbon stocks (Land C), split into vegetation (Veg C) and soil (Soil C) carbon, and gross primary productivity (GPP), net primary productivity (NPP) and conductance (g_s), between 1901 and 2050.

	GPP_hi ($Pg\ C\ yr^{-1}$)	GPP_low ($Pg\ C\ yr^{-1}$)	LandC_hi ($Pg\ C$)	LandC_low ($Pg\ C$)
Value in 1901:	9.05	9.34	167.00	167.50
Value in 2050:				
CO_2	11.63	11.84	184.22	187.07
O_3	8.25	8.94	155.35	162.84
$CO_2 + O_3$	10.84	11.49	174.83	182.35
† % change due to O_3 at PI CO_2	-8.84	-4.28	-6.98	-2.78
‡ % change due to O_3 at high CO_2	-6.79	-2.96	-5.10	-2.52
†† Alleviation of O_3 damage by CO_2 increase (%)	2.05	1.33	1.88	0.26

Table S6. Percentage reduction in simulated GPP and Land C by 2050 due to future O_3 effects at pre-industrial (PI) CO_2 concentration, and under increasing future CO_2 concentration. The difference between these defines the alleviation of the O_3 effect by CO_2. **O_3** = Fixed 1901 CO_2, Varying O_3 ; **CO_2** = Varying CO_2, Fixed 1901 O_3 ; **$CO_2 + O_3$** = Varying CO_2, Varying O_3. Calculated as: †) O_3 effect with fixed pre-industrial CO_2: 100*(fixCO_2_varO_3[2050] – value[1901])/value[1901], where value[1901] represents the hypothetical value at 2050 from a run with fixCO_2_fixO_3 which is equivalent to the initial state, i.e. the value in 1901 ; ‡) O_3 effect with increasing CO_2: 100*(varCO_2_varO_3[2050] - varCO_2_fixO_3[2050])/varCO_2_fixO_3[2050] ; ††) the alleviation of O_3 damage by CO_2 is the difference between the two runs: ‡ - †.

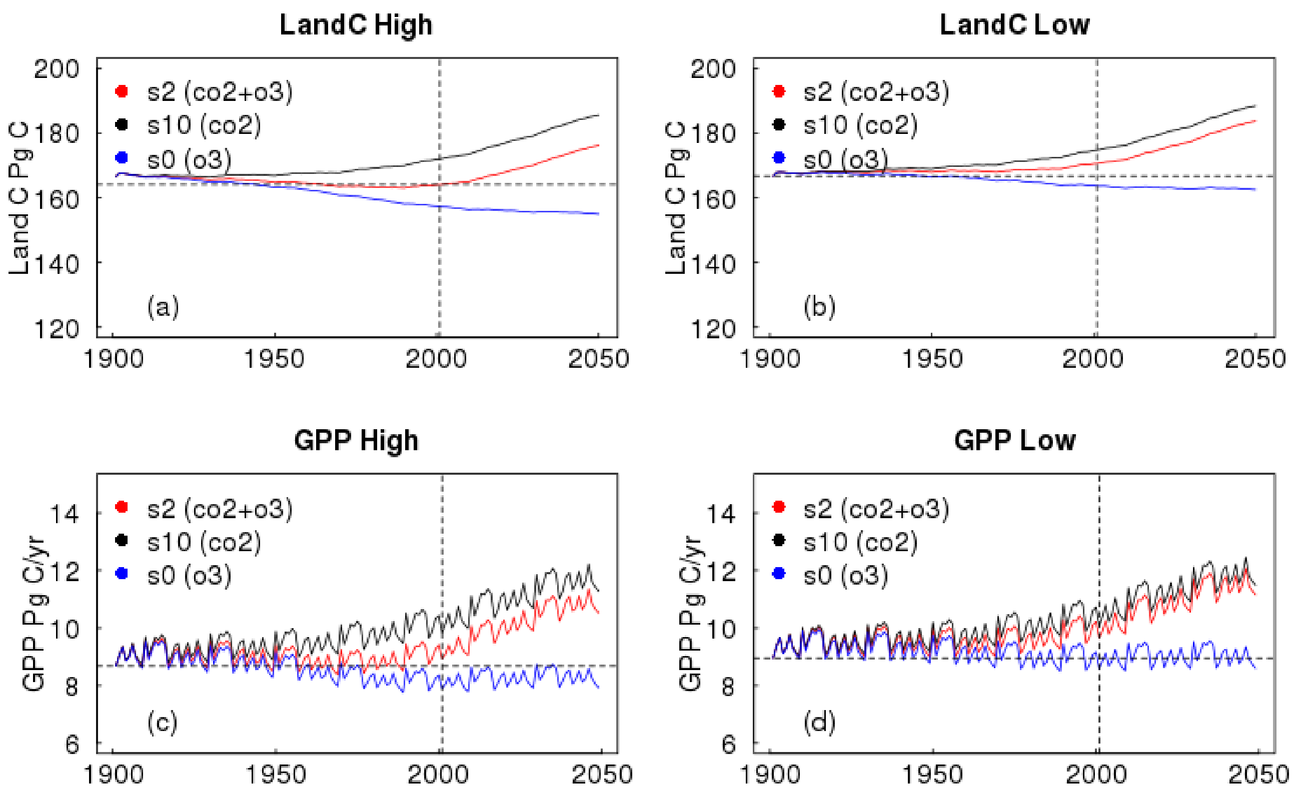

Figure S19. Times series (1901 to 2050) of changes in total carbon stocks (Land C) and gross primary productivity (GPP) due to O_3 effects at fixed pre-industrial atmospheric CO_2 concentration (O_3, blue), CO_2 effects at fixed pre-industrial O_3 concentration (CO_2, black), and effects of CO_2 and O_3 together (CO_2+O_3, red), for the higher and lower plant O_3 sensitivity. The horizontal dashed line shows the pre-industrial value, and the vertical dashed line marks the year 2001.

Acknowledgments

This work used eddy covariance data acquired and shared by the FLUXNET community, including these networks: AmeriFlux, AfriFlux, AsiaFlux, CarboAfrica, CarboEuropeIP, CarboItaly, CarboMont, ChinaFlux, Fluxnet-Canada, GreenGrass, ICOS, KoFlux, LBA, NECC, OzFlux-TERN, TCOS-Siberia, and USCCC. The ERA-Interim reanalysis data are provided by ECMWF and processed by LSCE. The FLUXNET eddy covariance data processing and harmonization was carried out by the European Fluxes Database Cluster, AmeriFlux Management Project, and Fluxdata project of FLUXNET, with the support of CDIAC and ICOS Ecosystem Thematic Center, and the OzFlux, ChinaFlux and AsiaFlux offices.