

1 **Ozone vegetation damage effects on gross primary productivity in the United States**

2
3 Xu Yue and Nadine Unger

4 School of Forestry and Environmental Studies, Yale University, 195 Prospect Street,
5 New Haven, CT 06511

6 Correspondence to: Xu Yue (xu.yue@yale.edu)

7
8 **Abstract**

9
10 We apply an off-line process-based vegetation model (the Yale Interactive Terrestrial
11 Biosphere model) to assess the impacts of ozone (O₃) vegetation damage on gross
12 primary productivity (GPP) in the United States during the past decade (1998-2007). The
13 model's GPP simulation is evaluated at 40 sites of the North American Carbon Program
14 (NACP) synthesis. The ecosystem-scale model version reproduces interannual variability
15 and seasonality of GPP at most sites, especially in croplands. Inclusion of the O₃ damage
16 impact decreases biases of simulated GPP at most of the NACP sites. The simulation
17 with the O₃ damage effect reproduces 64% of the observed variance in summer GPP and
18 42% on the annual average. Based on a regional gridded simulation over the U.S.,
19 summertime average O₃-free GPP is 6.1 g C m⁻² day⁻¹ (9.5 g C m⁻² day⁻¹ in the East of
20 95°W and 3.9 g C m⁻² day⁻¹ in the West). O₃ damage decreases GPP by 4-8% on average
21 in the eastern U.S. and leads to significant decreases of 11-17% in east coast hotspots.
22 Sensitivity simulations show that a 25% decrease in surface O₃ concentration halves the
23 average GPP damage to only 2-4% suggesting the substantial co-benefits to ecosystem
24 health that may be achieved via O₃ air pollution control.

25
26 Keywords: Ozone, gross primary productivity, photosynthesis, stomatal conductance

31 **1 Introduction**

32

33 The effects of tropospheric ozone (O₃) damage on U.S. forests have been studied for half
34 a century (Karnosky et al., 2007), but the impacts of O₃ on the North American carbon
35 balance are still relatively poorly understood (Felzer et al., 2004; Huntingford et al.,
36 2011). O₃ is a secondary pollutant produced in the troposphere during the photochemical
37 oxidation of carbon monoxide, methane, and volatile organic compounds (VOCs) by the
38 major tropospheric oxidant, the hydroxyl radical, in the presence of sunlight and nitrogen
39 oxides. Fossil-fuel, biofuel and biomass burning since the industrial and agricultural
40 revolutions have greatly increased the emissions of O₃ precursors and led to an
41 approximate doubling of O₃ levels over the U.S. since the preindustrial. Deposition
42 through stomatal uptake is an important sink for O₃ but damages photosynthesis, reduces
43 plant growth and biomass accumulation, limits crop yields, and affects stomatal control
44 over plant transpiration of water vapor between the leaf surface and atmosphere
45 (Ainsworth et al., 2012; Hollaway et al., 2012).

46

47 Understanding the O₃ pollution influence on the North American forest sink is crucial to
48 any effort to mitigate climate change by stabilizing atmospheric carbon dioxide (CO₂)
49 concentrations. Currently, North America is acting as a net source of CO₂ to the
50 atmosphere (King et al., 2012). Sequestration of atmospheric CO₂ by forest ecosystems is
51 a major control on atmospheric CO₂ abundance and its growth rate (Pan et al., 2011).
52 Terrestrial ecosystems of North America absorb the equivalent of about 35% of North
53 America's fossil fuel based CO₂ emissions, representing a source-to-sink ratio of nearly
54 3:1. Forest growth in the U.S. is responsible for 30–70% of this North American CO₂
55 sink, which varies significantly from year to year (Pacala et al., 2001; Goodale et al.,
56 2002; Pan et al., 2011; King et al., 2012). Worse still, there is evidence that the summer
57 continental U.S. is more sensitive than other world regions to climate forcing (Levy et al.,
58 2008).

59

60 Experimental studies that examine O₃ impacts on plant productivity are typically
61 performed for individual vegetation types, on the scale of sites, and within a limited time

62 period (e.g. Wittig et al., 2007; Feng et al., 2008; Lombardozzi et al., 2013). For example,
63 based on measurements reported from over 100 studies, Wittig et al. (2007) estimated
64 that chronic O₃ exposure depressed photosynthesis by 11% and stomatal conductance by
65 13% for several tree species at the ambient O₃ level of ~45 ppbv relative to that in O₃-
66 free air. The O₃ damage effect is strongest for crops. With datasets from ~50 peer-
67 reviewed studies, Feng et al. (2008) estimated that elevated O₃ levels significantly
68 decrease wheat photosynthetic rates by 20% and stomatal conductance by 22%.
69 Emerging research has found that the O₃ vegetation damage effects may result in a loss of
70 plant stomatal control, and a consequent decoupling of the stomatal response from
71 photosynthesis inhibition (Lombardozzi et al., 2012a, 2012b, 2013).

72

73 Previous work has found that in the U.S. region during 1989-1993, O₃ pollution reduced
74 net primary productivity (NPP) by 3-7% overall, and up to 13% in hotspots including the
75 southeast and in the Midwest agricultural lands (Felzer et al., 2004, 2005). The indirect
76 CO₂ radiative forcing due to the vegetation damage effects of anthropogenic O₃ increases
77 since the industrial revolution may be as large as +0.4 Wm⁻² (Sitch et al., 2007), which is
78 25% of the magnitude of the direct CO₂ radiative forcing over the same period, and of
79 similar magnitude to the direct O₃ radiative forcing. Through this perturbation of the
80 carbon cycle, O₃ pollution affects the climate system on considerably longer timescales
81 than its own atmospheric lifetime (Unger and Pan, 2012). Over the past decade since this
82 previous assessment surface O₃ levels in most of the U.S. have decreased (Lefohn et al.,
83 2010) due to domestic emission reductions following the implementation of air quality
84 control legislation (Bloomer et al., 2010). However, increasing O₃ concentration is
85 observed over western U.S. (Jaffe and Ray, 2007). Such a trend may in part be related to
86 the inter-continental flow from Asia (Cooper et al., 2010) and the global increase in
87 methane (Rigby et al., 2008).

88

89 The major goal of this study is to assess O₃ damage effects on gross primary productivity
90 (GPP) in the U.S. for the recent decade 1998-2007 using a data-constrained vegetation
91 model. In this work, we describe the implementation of a semi-mechanistic O₃ damage
92 function (Sitch et al., 2007) into the **Yale Interactive Terrestrial Biosphere** model (YIBs)

93 that includes enzyme-kinetic biophysics (Unger et al., 2013). In the first stage of the
94 study, we utilize eddy-derived GPP flux measurements at 40 sites across the U.S. and
95 Canada that have been collated for the North American Carbon Program (NACP) site-
96 level interim synthesis (Huntzinger et al., 2012; Schaefer et al., 2012; Barr et al., 2013;
97 Ricciuto et al., 2013) to evaluate an off-line version of the vegetation model's site level
98 GPP simulation and to assess the impact of surface O₃ damage at those sites. In the
99 second stage of the study, the impacts of O₃ damage on GPP throughout the entire U.S.
100 region are quantified using a regionally distributed configuration of the vegetation model.

101

102 **2 Methodology and data**

103

104 **2.1 Vegetation biophysics**

105

106 Here, we apply an off-line version of the YIBs model that previously was implemented
107 into the NASA Goddard Institute for Space Studies global chemistry-climate model
108 (Unger et al., 2013). The off-line model can be run at the site-level or in distributed mode
109 for a designated region. The vegetation biophysics module computes the photosynthetic
110 uptake of CO₂ coupled with the transpiration of water vapor at the 1-hour physical
111 integration time step of the off-line model. The vegetation biophysics calculates C3 and
112 C4 photosynthesis using the well-established Michaelis-Menten enzyme-kinetics leaf
113 model of photosynthesis (Farquhar et al., 1980; von Caemmerer and Farquhar, 1981) and
114 the stomatal conductance model of Ball and Berry (Collatz et al., 1991). The coupled
115 photosynthesis/stomatal conductance leaf model has been widely used to project
116 terrestrial biosphere responses to global change. The model is briefly summarized here
117 for transparency and completeness. The leaf model assumes that the rate of net CO₂
118 assimilation (A_{net}) in the leaves of C3 and C4 plants is limited by one of three processes:
119 (i) the capacity of the ribulose 1,5-bisphosphate (RuBP) carboxylase-oxygenase enzyme
120 (Rubisco) to consume RuBP (J_c); (ii) the capacity of the Calvin cycle and the thylakoid
121 reactions to regenerate RuBP supported by electron transport (J_e); (iii) the capacity of
122 starch and sucrose synthesis to consume triose phosphates and regenerate inorganic
123 phosphate for photo-phosphorylation in C3 and phosphoenolpyruvate (PEP) limitation in

124 C4 (J_s). J_c , J_e , and J_s are described as functions of the maximum carboxylation capacity
 125 (V_{cmax}) at the optimal temperature, 25°C, and the internal leaf CO₂ concentration (C_i). The
 126 gross rate of carbon assimilation from photosynthesis (A) is given by:

$$127 \quad A = \min(J_c, J_e, J_s) \quad (1)$$

128 Net carbon assimilation is given by:

$$129 \quad A_{net} = A - R_d \quad (2)$$

130 where R_d is the rate of dark respiration:

$$131 \quad R_d = 0.015 \cdot V_{cmax} \quad (3)$$

132 Leaf stomata control the uptake of CO₂ versus the loss of H₂O. At equilibrium, the
 133 stomatal conductance of water vapor through the leaf cuticle (g_s in mol [H₂O] m⁻² s⁻¹)
 134 depends on the net rate of carbon assimilation:

$$135 \quad g_s = m \frac{A_{net} \cdot RH}{c_s} + b = \frac{1}{r_s} \quad (4)$$

136 where m and b are the slope and intercept derived from empirical fitting to the Ball and
 137 Berry stomatal conductance equations, RH is relative humidity, c_s is the CO₂
 138 concentration at the leaf surface, and r_s is the stomatal resistance to water vapor.
 139 Appropriate photosynthesis parameters for the local vegetation type are taken from
 140 (Friend and Kiang, 2005) and the Community Land Model (Oleson et al., 2010) with
 141 updates from Bonan et al. (2011) (Table 1). In both the site-level and distributed models,
 142 we apply these model PFT-specific photosynthesis parameters and do not tune or
 143 calibrate to the local vegetation properties. The model calculates evapotranspiration as a
 144 function of the stomatal conductance. However, we do not consider the feedback of the
 145 changes in evapotranspiration to the boundary-layer meteorology because we use
 146 prescribed meteorological variables from reanalyses in the simulations.

147

148 The canopy radiative transfer scheme assumes a closed canopy and layers the canopy for
 149 light stratification using an adaptive number of layers (typically 2-16) (Friend and Kiang,
 150 2005). Each canopy layer distinguishes sunlit and shaded regions for which the direct and
 151 diffuse photosynthetically active radiation (PAR) is computed (Spitters et al., 1986). The
 152 coupled photosynthesis and stomatal conductance equations are solved analytically using
 153 a cubic function of A_{net} . C_i is calculated explicitly at the leaf level. Scaling of the leaf to

154 canopy level is through stratification of canopy light levels and leaf area profiles. The
 155 photosynthetic uptake of CO₂ is accumulated into a carbon reserve pool, from which
 156 other processes may allocate uses.

157

158 **2.1.1 O₃ damage effect on photosynthesis**

159

160 O₃ oxidizes cellular membranes and photosynthetic tissues when it enters leaves through
 161 stomata, leading to reductions in photosynthesis and GPP. O₃ damage inhibits stomatal
 162 conductance, which is closely related to the photosynthetic rate, resulting in a reduction
 163 in transpiration. A semi-mechanistic parameterization is employed to estimate the O₃
 164 damage effects to both photosynthesis and stomatal conductance (Sitch et al., 2007). The
 165 exposure to O₃ leads to reductions in photosynthesis:

$$166 \quad A' = F \cdot A_{net} \quad (5)$$

167 where F is the reduction fraction calculated as

$$168 \quad F = 1 - a \cdot U_{>O3T} \quad (6)$$

169 where a is the O₃ sensitivity coefficient derived from observations. Two cases are
 170 examined: high and low O₃ sensitivity following Sitch et al. (2007). $U_{>O3T}$ is the
 171 instantaneous leaf uptake of O₃ flux above a plant function type (PFT)-specific threshold
 172 of O3T (Table 1),

$$173 \quad U_{>O3T} = \max[(F_{O3} - O3T), 0] \quad (7)$$

174 here F_{O3} is the O₃ flux entering the leaf through the stomata,

$$175 \quad F_{O3} = \frac{[O_3]}{r_b + \kappa \cdot r_s'} \quad (8)$$

176 where $[O_3]$ is the O₃ concentration at the top of the canopy, r_b is the boundary layer
 177 resistance. The stomatal resistance to O₃ is calculated based on stomatal resistance to
 178 water r_s with a ratio constant $\kappa=1.67$. From Equation (4), the decrease in A_{net} reduces the
 179 stomatal conductance g_s proportionally,

$$180 \quad r_s' = \frac{1}{g_s'} = \frac{1}{F \cdot g_s} \quad (9)$$

181 The r_s' and g_s' are the O₃-damaged stomatal resistance and conductance, respectively.
182 When the plant is exposed to [O₃] (Equation 8), the excess O₃ flux entering leaves
183 (Equation 7) causes $F < 1$ (Equation 6), decreasing A_{net} (Equation 5) while increasing the
184 stomatal resistance (Equation 9). The latter will act to reduce the O₃ uptake flux
185 (Equation 8) to protect the plant. Thus, the scheme considers associated changes in both
186 photosynthetic rate and stomatal conductance. When photosynthesis is inhibited by O₃,
187 the stomatal conductance decreases accordingly to resist more air passing through the
188 stomata, resulting in a decline of the oxidant fluxes inside leaves, as described through
189 equations (5)-(9). Consequently, this coupled scheme represents the equilibrium state
190 between the CO₂ demand for vegetation growth and the protection against O₃ damage by
191 plant. The parameters for the scheme, including the O₃ damage threshold and sensitivity
192 coefficients, were originally derived based on the calibration of the MOSES vegetation
193 model. Since the MOSES model employs the (almost) identical Farquhar-Ball-Berry
194 photosynthesis/stomatal conductance scheme as in the YIBs model, it is appropriate to
195 adopt the same parameters as those derived in Sitch et al. (2007) (Table 1). Evaluation of
196 the YIBs simulated O₃-induced GPP response with available field and laboratory
197 measurements across a range of PFTs in Section 3.4 indicates that our assumption is
198 reasonable.

199

200 **2.1.2 Vegetation structure**

201

202 The YIBs vegetation model simulates eight PFTs, using either C3 or C4 photosynthesis
203 (Table 1). We apply two different sets of land cover and leaf area index (LAI) in the
204 simulations. The first set is the PFT-specified vegetation cover fraction and LAI retrieved
205 by the Moderate Resolution Imaging Spectroradiometer (MODIS, Knyazikhin et al.,
206 1998). The value on a specific day is linearly interpolated from the monthly means of the
207 nearest two months based on the distance of this day to the middle dates of those two
208 months. The second set uses LAI from the Global Modeling and Assimilation Office
209 (GMAO) Modern Era-Retrospective Analysis (MERRA) dataset. The MERRA LAI is
210 assimilated based on radiance data retrieved by over 20 satellites (Rienecker et al., 2011)
211 and is available on daily scale from 1980 onwards. Since the MERRA LAI dataset does

212 not provide PFT-specific information, the actual site-level PFT is assumed for the site
213 level simulations. For the regional distributed simulations, the land cover is prescribed to
214 the gridded International Satellite Land-Surface Climatology Project (ISLSCP, Hall et al.,
215 2006).

216

217 **2.1.3 Meteorological forcing**

218

219 For the site-level simulations, we use hourly *in situ* measurements of surface
220 meteorological variables, including surface air temperature, specific humidity, wind
221 speed, surface pressure, and CO₂ concentrations. There are some missing values in the
222 measurements due to occasional instrument failure. We gap-fill the site-based
223 observations with that from the MERRA-land data (Reichle et al., 2011), which is
224 interpolated to each site based on the site location.

225

226 For the distributed simulations, the off-line YIBs model uses hourly MERRA-land data
227 climatic variables including: surface air temperature, specific humidity, wind speed,
228 surface pressure, precipitation, direct PAR, and diffuse PAR, and soil temperature and
229 soil moisture at 6 soil depths. The original data resolution of 0.5°×0.667° by latitude and
230 longitude is degraded to 1°×1.333° due to current disk space limitation.

231

232 **2.1.4 Surface [O₃]**

233

234 Hourly and daily maximum 8-hour average surface [O₃] representative of the present day
235 climate (~ 2005) are taken from previous simulations using NASA Model-E2 (Shindell et
236 al., 2013). The global model has 2°×2.5° latitude by longitude horizontal resolution with
237 40-vertical layers extending to 0.1 hPa. The gas-phase chemistry and aerosol modules are
238 fully integrated, so that these components interact with each other and with the physics of
239 the climate model (Bell et al., 2005; Shindell et al., 2006; Unger, 2011; Shindell et al.,
240 2013). The model surface O₃ is validated using measurements from 73 Clean Air Status
241 and Trends Network (CASTNET) sites operated by the United States Environmental
242 Protection Agency (EPA) (<http://epa.gov/castnet/javaweb/ozone.html>) and ~1200

243 monitor sites managed by the EPA AIRDATA (<http://www.epa.gov/airdata/>). These sites
244 are operated on the county level scale. The CASTNET provides hourly [O₃] at rural sites
245 from 1996-2005. The AIRDATA network provides daily maximum 8-hour average
246 (MDA8) [O₃], covering both urban and rural regions. We use AIRDATA data for the
247 year 2005.

248

249 **2.2 Simulations**

250

251 **2.2.1 Site-level runs**

252

253 We configure a site-level version of the YIBs model for the 40 eddy covariance flux
254 tower sites described in detail in the NACP synthesis (Fig. S1 and Appendix Table A,
255 Schaefer et al., 2012). Measurements are available for a wide range of time periods
256 across the different sites ranging from the minimum of 1 year at Fermi Lab (US-IB1) and
257 the maximum of 15 years at Harvard Forest (US-HA1). These sites cover a range of
258 different vegetation types including: evergreen needleleaf forest, deciduous broadleaf
259 forest, grasslands, croplands, closed shrublands, mixed forests, permanent wetlands, and
260 woody savannas. Table S1 summarizes how the NACP vegetation types are mapped onto
261 the 8 model PFTs. For the site-level simulations, we assume C4 photosynthetic pathway
262 for all cropland sites, which are mainly corn (Schaefer et al., 2012).

263

264 For each site, a group of six sensitivity simulations are performed (Table 2). We conduct
265 the first four runs using different combinations of meteorological and vegetation forcings,
266 to assess the sensitivity of the results to local versus reanalysis meteorological forcing and
267 LAI (Table 2). Two, METmerra_LAImodis and METmerra_LAI merra, use hourly
268 meteorology from MERRA-land reanalyses alone. The other two, METsite_LAI modis
269 and METsite_LAI merra, use site-based meteorology with gap-filled MERRA reanalysis.
270 Simulations use two datasets of LAI: (1) METmerra_LAI merra and METsite_LAI merra
271 use LAI from the MERRA-land reanalyses, which provide non PFT-specific LAI that we
272 assign to the local PFT type at each site (Table A), while (2) METmerra_LAI modis and
273 METsite_LAI modis use PFT-specific LAI retrieved by the MODIS. We perform two

274 additional site-level simulations, which use the same forcings as that for
275 METsite_LAImerria but with the impact of O₃ uptake on photosynthesis. These two
276 experiments, METsite_LAImerria_HO3 and METsite_LAImerria_LO3, use either high or
277 low O₃ sensitivity as defined by the coefficient *a* in Table 1.

278

279 To quantify the performance of the vegetation model, we estimate the χ^2 for each site
280 following the method described in Schaefer et al. (2012),

$$281 \quad \chi^2 = \frac{1}{n} \sum_{i=1}^n \left(\frac{r_i}{\varepsilon_i} \right)^2 \quad (10)$$

282 where

$$283 \quad r_i = (GPP_{si} - GPP_{oi}) \quad (11)$$

284 is the difference between the pair of simulated and observed GPPs. ε_i are the
285 observation uncertainties. *n* is the length of observations (e.g. the number of days for
286 the daily variables). The lower the χ^2 , the smaller the model biases. If $\chi^2 < 1$, the
287 simulation bias is on average smaller than the measurement uncertainty, indicating a
288 good performance of the model. Here, we define a reasonable performance of $\chi^2 < 4$,
289 when the residual is less than twice the measurement uncertainty. We also calculate the
290 root mean square error (RMSE) as follows:

$$291 \quad RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (GPP_{si} - GPP_{oi})^2} \quad (12)$$

292


293 We validate the simulated O₃ damage effect with measurements from literature. Field and
294 laboratory experiments may have different [O₃] compared to the ambient level we used
295 complicating the validation. As a result, we perform 14 additional sensitivity simulations
296 for each of NACP sites. All tests use meteorological and vegetation forcings the same as
297 METsite_LAImerria (Table 2), except for the different [O₃] and O₃ sensitivity. These
298 experiments are divided into two groups, 7 in each, using either low or high O₃
299 sensitivity. In each group, simulations are performed with constant [O₃] at 20, 40, 60, 80,
300 100, 120, 140 ppbv, respectively. We do not include diurnal and seasonal variations of
301 [O₃] in these sensitivity simulations as that in METsite_LAImerria for two reasons. First,

302 field measurements for the O₃ vegetation damage are usually performed with fixed [O₃]
303 during the growth season (e.g. Ishii et al., 2004; Zhang et al., 2012). Second, the diurnal
304 cycles and seasonality of [O₃] are very different for different sites (Bloomer et al., 2010),
305 making it difficult to apply a uniform temporal cycle for all the NACP sites. The
306 reductions in GPP at these simulations are compared with results from field
307 measurements at the corresponding [O₃] level.

308

309 **2.2.2 Distributed run over U.S. region**

310

311 A gridded version of the YIBs model at 1°×1.333° latitude by longitude horizontal
312 resolution for the U.S. region is driven with MERRA meteorological forcings for the
313 period 1998-2007. In the distributed model, vegetation cover types are from the ISLSCP
314 and LAI is from the MERRA-land reanalysis. We assign the MERRA LAI to the
315 corresponding PFT types defined by ISLSCP (Fig. S2). The 18 ISLSCP land types are
316 converted to 8 PFTs used in the model (Table S1). Some of the ISLSCP land types, such
317 as the deciduous needleleaf forest, are not represented in the YIBs model. However, the
318 coverage of these types is very small in the U.S. (Fig. S2) and will not influence the
319 regional simulation after the conversion to the model types. For the regional simulation,
320 we assume that the total crop area in each crop grid cell is split 50% C3 and 50% C4 to
321 account for the dominance of both soybean (C3) and corn (C4) crops in the central and
322 northern U.S. agricultural regime. We perform 2 simulation cases with high and low O₃
323 damage sensitivity. Finally, to understand how the O₃ vegetation damage effect may
324 respond to possible future changes in [O₃], we perform four additional sensitivity
325 experiments with ±25% changes in [O₃] for each O₃ sensitivity 

326

327 **3. Results**

328

329 **3.1 Evaluation of O₃-free GPP at NACP sites**

330

331 We compare the monthly mean LAI from MERRA and MODIS at each NACP site (Fig.
332 1). For each site, the MERRA LAI is averaged for the period when GPP measurements

333 are available. The two datasets show similar annual cycles at several sites but are
334 inconsistent for 7 out of 20 evergreen sites (CA-Ca1, CA-Ca2, CA-Ca3, CA-NS1, US-
335 Me2, US-Me3, and US-Me5) and 2 out of 5 shrubland sites (US-SO2 and US-Ton). In
336 addition, for grasslands and croplands, the datasets exhibit different seasonality, with
337 MERRA LAI near zero during the cold season in contrast to the MODIS LAI that is
338 positive all year round. It must be emphasized that the MERRA and MODIS LAI
339 represent the average state in the retrieval product grid cells and as such may not
340 represent the local LAI for the actual PFT species at each site. The local site LAI values
341 are not available for comparison. For example, for most of the evergreen NACP sites, the
342 MERRA and MODIS LAI indicate large seasonality.

343

344 The long-term monthly mean O₃-free GPP from the simulation METsite_LAI_{merra} is
345 compared with observations at individual NACP sites (Fig. 2). Both the observation and
346 simulation are averaged over the measurement period at each site. The simulations
347 capture the magnitude and seasonality of GPP for most sites especially for deciduous
348 broadleaf and shrubland. The largest model overestimate (factor of 3-8) occurs at CA-SJ1
349 and CA-SJ2. For the grassland sites US-ARM (in Great Plains) and US-Var (in
350 California), the model maximum GPP occurs in summer (July), 2-3 months later than in
351 the measurements (April). This incorrect model seasonality is a result of the MERRA
352 LAI (compare Fig. 1) that does not begin to increase rapidly until after May and is not
353 consistent with the local LAI at the site. In reality, California grasslands exhibit rapid
354 growth in spring then mature and die after April or May (Chiariello, 1989). The
355 grasslands in the Great Plains may have up to six different phenological groups, including
356 some species active in spring (e.g. in US-ARM) while some others peak in summer (e.g.
357 in US-Shd) (Henebry, 2003). The correlation coefficient between simulated annual mean
358 GPP and observations at all 40 sites is 0.65. The correlation is higher (0.81) for summer
359 (June-August, Fig. S3). The annual GPP averaged over all sites is 3.8 g C m⁻² day⁻¹, 27%
360 higher than the observational average (3.0 g C m⁻² day⁻¹).

361

362 Among the 40 NACP sites, 22 have reasonable performance with $\chi^2 < 4$ (Fig. 3a). The
363 simulation is the best ($\chi^2 = 1.2$) at the evergreen needleleaf site US-Dk3 (Fig. 2). For the

364 top 10 sites with the lowest biases, 8 are based in evergreen needleleaf forests. These
365 sites usually have multiple years of measurements and provide good samples for testing
366 the consistency between simulations and observations. Simulations at 4 broadleaf forest,
367 4 cropland, and 3 shrubland sites have $\chi^2 < 4$, and the latter usually has low GPP whose
368 peak is not higher than $10 \text{ g C m}^{-2} \text{ day}^{-1}$ (Fig. 2). Compared with the 24 land surface
369 models in Schaefer et al. (2012), the YIBs model shows significant improvement at the
370 crop PFT sites ($\chi^2 < 4.1$ vs. $\chi^2 > 6$). YIBs simulates GPP with $\chi^2 < 4$ at 22 sites in total
371 compared to 16 sites for the ensemble simulations in Schaefer et al. (2012). YIBs GPP
372 simulation is weaker ($\chi^2 > 4$) at 18 sites including 9 needleleaf forest, 2 broadleaf, and 2
373 shrubland PFTs. The common feature of the biases at these sites is the overestimation of
374 peak GPP during summer (e.g. CA-SJ1, CA-SJ2, CA-Mer, Fig. 2). It is possible that the
375 model does not represent the full realism of the biophysical processes accurately.
376 However, we assert that the most likely cause of the model overestimate is the uniform
377 application of model PFT-specific photosynthesis parameters that are not tuned to local
378 site level vegetation parameters and, for instance, do not take into account plant
379 species and age. Similar to the multi-model results in Schaefer et al. (2012), YIBs
380 performance is weakest at the 5 grassland sites. In this case, the bias is mainly due to the
381 delayed LAI seasonality in the MERRA satellite dataset (Figs. 1 and 2). In general,
382 application of the remotely sensed LAI is a source of error because the gridded satellite
383 data may not represent the local site changes in plant growth and phenology, especially
384 for vegetation types with low biomass. The limitation of the satellite LAI spatial
385 resolution implies that the model is unable to resolve GPP variability for sites in close
386 proximity. For example, sites CA-SJ1, CA-SJ2, and CA-SJ3 are located close to each
387 other. Simulations at these sites have similar magnitude in simulated GPP while
388 observations show distinct variability between the sites.

389

390 We compare R^2 , RMSE, and χ^2 for the different sensitivity experiments in order to
391 ascertain which combination of meteorological and LAI forcings best reproduces the
392 measured GPP over North America (Table 3 and Fig. 4). CA-Let, CA-NS1, US-Var, CA-
393 SJ1, and CA-SJ2 are excluded from the analysis because of the excessive bias at those
394 sites (Fig. 3a). The average R^2 increases while RMSE decreases when MERRA

395 reanalyses are substituted with site-based meteorology, or the MERRA LAI is used
396 instead of MODIS LAI (Table 3). The choice of LAI forcing has the most significant
397 impact on YIBs simulation performance consistent with previous work that demonstrated
398 the dominance of phenology over meteorology in controlling local terrestrial carbon
399 exchange (Desai et al., 2008; Puma et al., 2013). Using MODIS LAI, YIBs has a total χ^2
400 of 9.2 that shows an average reduction of 4.7 (52%) with MERRA LAI (Table 3 and Fig.
401 4). Applying the site meteorology relative to MERRA meteorological forcings offers
402 smaller improvements. For example, the total χ^2 value decreases by 5% in
403 METsite_LAI_{modis} compared with that in MET_{merra}_LAI_{modis} and 15% in
404 METsite_LAI_{merra} relative to that in MET_{merra}_LAI_{merra} (Table 3).

405

406 **3.2 Evaluation of modeled surface [O₃]**

407

408 We validate summertime surface O₃ simulated by the NASA Model-E2 chemistry-
409 climate model with observations from the CASTNET and AIRDATA (Fig. 5). High O₃
410 level appears in the eastern U.S. due to anthropogenic emissions and in the mountainous
411 western U.S. due to high elevation. The model generally captures this spatial pattern with
412 a correlation coefficient of 0.39 against observations over the selected 73 CASTNET
413 sites (Figs. 5a-b). The simulation overestimates the O₃ level by ~4 ppbv (12%) in the East
414 and ~1 ppbv (3%) in the West. The CASTNET sites are located in rural sites, which
415 usually have lower [O₃] than that in urban areas, except for some megacities where the
416 excessive NO_x emissions result in lower O₃ level (Gregg et al., 2003). Therefore, we also
417 compare the simulated MDA8 [O₃] with monitored at ~1200 AIRDATA sites, which
418 covers both urban and rural regions (Fig. 5c). In the eastern U.S., the model captures high
419 [O₃] centers around Michigan, Indiana, and Ohio states and that along the northeast coast.
420 In the West, the simulation reproduces high [O₃] over mountain regions and in California.
421 On average, the simulated MDA8 [O₃] is lower by ~0.5 ppbv (1%) in the East and ~3.5
422 ppbv (7%) in the West. The correlation coefficient between simulations and observations
423 is as high as 0.51 (Fig. 5d).

424

425 **3.3 O₃ damage effects at NACP sites**

426

427 We must apply the simulated O₃ to quantify the O₃ vegetation damage at the NACP sites
428 because the sites do not monitor local [O₃]. The summer average [O₃] is 30-50 ppbv at 24
429 U.S. sites (Fig. 6a). The O₃ damage effect is relatively stronger at sites with both high O₃-
430 free GPP and ambient [O₃] (Fig. 6d). The most significant damages are predicted at US-
431 MMS (DBF) and US-Dk3 (ENF) sites, where the GPP reductions are 5-14% depending
432 on the low or high O₃ sensitivity (Fig. 6d). At these two sites, the high stomatal
433 conductance (4.0 and 3.4 mm s⁻¹, Fig. 6b) and ambient [O₃] (both 43 ppbv, Fig. 6a) result
434 in the largest O₃ stomatal flux (both ~0.3 mmol m⁻² d⁻¹, Fig. 6c) among the 24 sites. The
435 lowest O₃ damage (1-2% GPP reduction) appears in the 3 shrub sites, US-Ton, US-SO2,
436 and US-Los, although mean [O₃] there is as high as 43 ppbv. The main reason for the
437 limited O₃ damage is the low stomatal conductance (average 1.4 mm s⁻¹, Fig. 6b) related
438 to the small O₃-free GPP (average 4.6 g C m⁻² d⁻¹, Fig. 6d). Similarly, the O₃ damage for
439 C3 grass is as low as 1-2%, although the GPP of this plant is highly sensitive to O₃ (Table
440 1). For needleleaf and broadleaf forest sites, the average site-level O₃ damage effects are
441 estimated to be 2-5% and 3-9% respectively with differences between these ecosystem
442 types predominantly driven by differences in sensitivity to O₃. The four C4 crop sites,
443 US-Ne1, US-IB1, US-Ne2, and US-Ne3, exhibit the highest O₃-free GPP but show only
444 moderate O₃ damage effects (GPP reductions of 4-6%, Fig. 6d). This result is driven by
445 low ambient [O₃] at the C4 crop sites (average 32 ppbv, Fig. 6a) in combination with the
446 reduced C4 stomatal conductance (higher water use efficiency) relative to C3 plants
447 (average 3.2 mm s⁻¹, Fig. 6b). Indeed, the C4 photosynthetic pathway has been observed
448 to offer protection against O₃ damage (Heagle et al., 1989; Rudorff et al., 1996).

449

450 Inclusion of O₃ damage effect improves the site-level simulations (Figs. 3b-c). For 36 out
451 of the 40 sites, the χ^2 of simulated GPP decreases when considering vegetation responses
452 to O₃, and the improvement is better when higher O₃ sensitivity is applied. At these sites,
453 for example, CA-TP4, US-Dk3, US-MMS, and US-PFa, the reduced GPP at peak seasons
454 is closer to measurements (Fig. S4), leading to smaller biases for simulations. On
455 average, the χ^2 decreases by 3-8% at these sites, depending on the O₃ sensitivity in the
456 simulation (Fig. 4). Finally, the simulated annual GPP averaged over all NACP sites

457 changes from $3.8 \text{ g C m}^{-2} \text{ day}^{-1}$ to $3.6 \text{ g C m}^{-2} \text{ day}^{-1}$ with the high O_3 sensitivity
458 simulation case, closer to the observations of $3.0 \text{ g C m}^{-2} \text{ day}^{-1}$. The bias-correction from
459 O_3 damage is much smaller relative to the effect of phenology (Fig. 4). Moreover, the O_3 -
460 induced damage does not improve the GPP correlation between observations and
461 simulations, which remains similar at ~ 0.8 (for 40 sites) with and without O_3 effects (Fig.
462 S3).

463

464 **3.4 Evaluation of simulated O_3 vegetation damage against field and laboratory data**

465

466 We compare the simulated O_3 damage effect with field and laboratory measurements
467 from the published literature (Fig. 7). In total, 14 additional sensitivity experiments are
468 performed with different levels of $[\text{O}_3]$ at each NACP site (see section 2.2.1). GPP
469 reductions increase accordingly with the increasing $[\text{O}_3]$ (Fig. 7). For a given $[\text{O}_3]$, the O_3
470 damage effect is strongest for C4 crops (despite the lower $g_s:A_{net}$ ratio) but weakest for
471 shrubland. YIBs simulates reasonable O_3 damage to GPP for all model PFTs compared to
472 the meta-analyses of Wittig et al. (2007) and Lombardozzi et al. (2013). Field studies in
473 shrubland are limited. Zhang et al. (2012) investigated the responses of four shrub species
474 to $[\text{O}_3]=70$ ppbv and found large reductions in net photosynthesis of 50-60%. The
475 average O_3 -free A_{net} of those shrub species was $8\text{-}16 \text{ g [C] m}^{-2} \text{ s}^{-1}$, much higher than even
476 the gross photosynthesis (A) of $6 \text{ g [C] m}^{-2} \text{ s}^{-1}$ at the shrub NACP sites, likely because the
477 latter are located in dry and/or cold areas (Fig. S1). The YIBs simulated O_3 vegetation
478 damage effects for C4 plants are in good agreement with field measurements from Taylor
479 et al. (2002) and Grantz et al. (2012). In the case of C3 grass and C3 crop, the model
480 simulates consistent GPP reduction percentages with observations from Feng et al. (2008)
481 for wheat, Foot et al. (1996) for *colluna vulgaris*, and Mulchi et al. (1992) for soybean.
482 However, these O_3 damage results are all $>50\%$ less than for available measurements in
483 rice crops (Ishii et al., 2004; Ainsworth et al., 2008), suggesting that rice may have much
484 higher O_3 sensitivity than other C3 plants. In the U.S. rice plantation area is much smaller
485 than that of soybean and corn. Therefore, we adopt the O_3 sensitivity parameters for
486 C3/C4 plants shown in Table 1 for the distributed regional simulations.

487

488 3.5 O₃ vegetation damage effect on GPP in U.S. region

489

490 High values of simulated summertime GPP (including O₃ damage effect) appear east of
491 95°W in the U.S. (Fig. 8a), because the land surface there is covered by crops and forests.
492 A high center of GPP (> 10 g C m⁻² day⁻¹) appears over cropland in the north central U.S.
493 In the western U.S., the coverage of grass and shrub and the low water availability (low
494 precipitation and soil moisture) over semi-arid regions lead to low carbon assimilation
495 rate. The regional gridded simulated GPP reproduces the JJA growing season average
496 NACP site-level fluxes with a correlation coefficient of 0.62 for 32 sites below 50°N
497 (Fig. 8a). The correlation is lower than the 0.84 estimated for the site-level simulation
498 METsite_LAI_{merra} at the same sites and the same season. Since the meteorological
499 forcings and LAI are similar, the difference in land cover, ISLSCP versus site definitions
500 (Fig. S1 and Fig. S2), accounts for the discrepancy between regional and site-level
501 simulations.

502

503 On average, the simulated summer GPP (including the high O₃ damage effect) is 9.5 g C
504 m⁻² day⁻¹ in the eastern U.S. and 3.9 g C m⁻² day⁻¹ in the western U.S., giving a mean
505 value of 6.1 g C m⁻² day⁻¹ for the U.S. region. The total carbon uptake is estimated to be
506 4.43 ± 0.18 Pg C during the summer growing season, accounting for 57-60% of the
507 annual average value of 7.59 ± 0.25 Pg C over the 1998-2007 period. Our estimate of
508 annual carbon uptake is consistent with previous published estimates. For example, Xiao
509 et al. (2010) upscaled site-level GPP flux to continental scale with a regression tree
510 approach based on both NACP fluxes and remote-sensing variables. They estimated that
511 the total GPP in U.S. ranges from 6.91 to 7.33 Pg C per year during 2000-2006. Using the
512 same observations but with a process-based biogeochemical model, Chen et al. (2011)
513 estimated a range of 7.02-7.78 Pg C per year for 2000-2005, which is even closer to our
514 estimate.

515

516 We calculate both O₃ stomatal flux (Fig. 8b) and the resultant damage on GPP (Fig. 9) in
517 the U.S. region for the 1998-2007 period. High O₃ stomatal flux is predicted in the
518 eastern U.S. due to co-location of the high GPP (medium to high stomatal conductance)

519 and the substantial ambient [O₃]. On average, the summertime O₃ plant uptake is 117
520 $\mu\text{mol m}^{-2} \text{ day}^{-1}$, with 207 $\mu\text{mol m}^{-2} \text{ day}^{-1}$ in the eastern U.S. and 59 $\mu\text{mol m}^{-2} \text{ day}^{-1}$ in the
521 western U.S. Following the O₃ stomatal flux, the largest mean GPP reductions are
522 predicted for the eastern U.S. growing season, in the range 4-8% depending on the O₃
523 sensitivity applied in the simulations (Fig. 9). Locally, reduction fraction reaches as high
524 as 11-17% in areas with high [O₃] pollution, such as Michigan, Indiana, Ohio, and states
525 along the northeast coast. Despite high surface [O₃] over mountainous elevated areas in
526 the West (Fig. 5), impacts on GPP are limited due to the low stomatal conductance and
527 low photosynthetic rate there. The Pacific northwestern forests are an exception, with a
528 moderate GPP reduction of 1-7%. On average, the total summer GPP is reduced by 2-5%
529 due to O₃ damage effects in the U.S. Similar reduction fractions are predicted for the
530 annual GPP.

531

532 U.S. surface [O₃] exhibits a decreasing trend over the past 2 decades, especially in the
533 eastern U.S., due to precursor emission controls (Lefohn et al., 2010). However, the
534 community continues to debate how surface [O₃] will respond to future emissions and
535 climate change. On the one hand, surface [O₃] may decline by the mid 21st century due to
536 large reductions in regional anthropogenic precursor emissions (Wu et al., 2008). On the
537 other hand, climate change effects alone may increase local surface [O₃] due to the
538 warmer, drier, and more stable environment (Leibensperger et al., 2008; Wu et al., 2008).
539 Due to the uncertainty in future surface [O₃] projections, our strategy here is to perform
540 four additional sensitivity experiments with $\pm 25\%$ changes in [O₃] for each O₃ sensitivity
541 case. Increases of 25% in [O₃] may reduce GPP in the eastern U.S. by 6-11%, with a
542 maximum local reduction of 25% for the high O₃ sensitivity case (Fig. 10d). The damage
543 magnitude with low O₃ sensitivity (Fig. 10b) mimics the present-day estimate with high
544 O₃ sensitivity (Fig. 9b). In contrast, the O₃ damage to the eastern U.S. GPP is as low as 2-
545 4% in response to 25% decreases in [O₃] (Figs. 10a, c), suggesting a substantial co-
546 benefit to ecosystem-health of O₃ precursor emissions control.

547

548 ~~4. Discussion and~~ Conclusions

549

550 We have performed an updated assessment of O₃ vegetation damage effects on GPP in
551 the U.S. for the 1998-2007 period using the YIBs vegetation model. The semi-
552 mechanistic parameterization of O₃ inhibition on photosynthesis proposed by Sitch et al.
553 (2007) has been implemented into this process-based vegetation model. The simulated O₃
554 damage effects are consistent with laboratory and field measurements reported in
555 previously published studies. We evaluated the simulated O₃-free and O₃-damaged GPP
556 with *in situ* measurements from 40 NACP sites. The O₃-free and O₃-damaged GPP
557 simulations capture the seasonality and interannual variability of GPP at most sites. The
558 model GPP biases are lowest at forest and cropland sites but highest at grassland sites.
559 Model GPP is highly sensitive to choice of LAI forcing. Simulations that apply MERRA
560 LAI generally perform better (show lower biases) than those with MODIS LAI. In
561 response to the simulated ambient [O₃] of 30-50 ppbv, simulated GPP decreases by 1-
562 14% at the NACP sites, depending on the O₃ sensitivity and PFT types. Maximum
563 reductions of 5-14% occur in two forest sites, where both O₃-free GPP and ambient [O₃]
564 are relatively high. Inclusion of the O₃ damage offers only a small improvement to the
565 simulated annual average GPP at NACP sites (from 3.8 g C m⁻² day⁻¹ to 3.6 g C m⁻² day⁻¹)
566 ¹) such that the model still overestimates the observational average of 3.0 g C m⁻² day⁻¹.
567 The model GPP overestimate is most likely related to the use of generic PFT-specific
568 photosynthesis parameters and the satellite prescribed LAI that may not represent the
569 local site LAI. In this work, we assumed a coupled response between photosynthesis and
570 stomatal conductance. Emerging research has found that the O₃ vegetation damage
571 effects can actually result in a loss of plant stomatal control, and a consequent decoupling
572 of the stomatal response from photosynthesis inhibition (Lombardozzi et al., 2012a, b).
573 Treatment of this decoupled response in the YIBs model would lead to a higher level of
574 O₃ flux entering leaves, thus causing stronger damage. Interestingly, this mechanism
575 would therefore provide a way to improve the simulated GPP overestimates. That said,
576 other studies have suggested that the O₃ damage effect is limited by carbon-nitrogen
577 interactions (Ollinger et al., 2002; Kvalevag and Myhre, 2013).
578
579 Regional distributed simulations for the U.S. yield a summertime GPP (with high
580 sensitivity O₃ damage) of 6.1 g C m⁻² day⁻¹ (9.5 g C m⁻² day⁻¹ in the eastern U.S. and 3.9

581 g C m⁻² day⁻¹ in the western U.S.). The total carbon uptake was estimated to be 4.43 ±
582 0.18 Pg C for the summer, accounting for 57-60% of the annual value of 7.59 ± 0.25 Pg
583 C over the 1998-2007 period. Carbon assimilation rate is suppressed by 4-8% on average
584 in the summertime eastern U.S. with maximum local damage of 11-17% in states close to
585 the Great Lakes and along the eastern coast. When [O₃] is decreased by 25%, O₃ damage
586 to GPP is only 2-4% in the eastern U.S., indicating substantial improvements to
587 vegetation health and carbon assimilation rate. Previously, Felzer et al. (2004) found
588 annual average O₃-induced NPP reductions of 3-7% over the U.S. for 1989-1993 and
589 simulated the largest reductions in states close to the Great Lakes and along the East
590 Coast, where the high O₃ sensitivity of crops makes the dominant contribution. Our study
591 examined O₃ damage effects a decade later than Felzer et al. (2004) but gives consistent
592 results. Qualitatively, this consistency between decades may be explained by the
593 offsetting influences of (i) surface O₃ reductions due to air quality control legislation and
594 (ii) GPP increases due to CO₂-fertilization and rising temperatures. Felzer et al. (2004)
595 estimated a maximum local NPP reduction of 34%, which is double the maximum of
596 17% in our analyses. Furthermore, Felzer et al. (2004) found widespread reductions of >
597 6% in the Midwest where there is almost no O₃ impact in this study (Fig. 9). Differences
598 between the studies are mostly likely driven by the use of different vegetation cover and
599 LAI datasets, and the use of a semi-mechanistic flux-based uptake in this study versus the
600 concentration-based uptake method elsewhere.

601

602 The current work has used an off-line approach. Yet, the O₃-vegetation-meteorology
603 system is strongly coupled. For instance, plant productivity itself controls the emission of
604 isoprene, a major O₃ precursor. The O₃-induced modification to stomatal conductance
605 may inhibit evapotranspiration, leading to changes in canopy temperature, precipitation,
606 soil moisture, and other surface hydrology and meteorology (Bernacchi et al., 2007;
607 vanLoocke et al., 2012). In future work, we will study O₃ vegetation damage effects
608 using YIBs embedded within a fully coupled global chemistry-climate model framework
609 in order to account for these feedbacks including altered canopy energy fluxes and
610 partitioning between latent and sensible heat that drive regional climate and hydrology. In
611 addition, future work will exploit recent extensive meta-data analyses (Lombardo et al.

612 al., 2013; Wittig et al., 2007) to refine the ozone damage parameterization in YIBs
613 including the decoupled modification of photosynthesis and stomatal conductance.

614

615 **Acknowledgements**

616 The authors are grateful to B. Felzer and an anonymous reviewer for assistance in
617 evaluating this paper. Data for this analysis was provided by the North American Carbon
618 Program Site Synthesis. Funding for this research was provided by Yale University. This
619 project was supported in part by the facilities and staff of the Yale University Faculty of
620 Arts and Sciences High Performance Computing Center.

621

622 **References**

- 623 Ainsworth, E. A.: Rice production in a changing climate: a meta-analysis of responses to
624 elevated carbon dioxide and elevated ozone concentration, *Global Change Biol.*, 14,
625 1642-1650, doi:10.1111/J.1365-2486.2008.01594.X, 2008.
- 626 Ainsworth, E. A., Yendrek, C. R., Sitch, S., Collins, W. J., and Emberson, L. D.: The
627 Effects of Tropospheric Ozone on Net Primary Productivity and Implications for
628 Climate Change, *Annu. Rev. Plant Biol.*, 63, 637-661, doi:10.1146/Annurev-Arplant-
629 042110-103829, 2012.
- 630 Barr, A., Ricciuto, D., Schaefer, K., Richardson, A., Agarwal, D., Thornton, P., Davis,
631 K., Jackson, B., Cook, R., Hollinger, D., Ingen, C. v., Amiro, B., Andrews, A., Arain,
632 M., Baldocchi, D., Black, T., Bolstad, P., Curtis, P., Desai, A., Dragoni, D., Flanagan,
633 L., Gu, L., Katul, G., Law, B., Lafleur, P., Margolis, H., Matamala, R., Meyers, T.,
634 McCaughey, H., Monson, R., Munger, J., Oechel, W., Oren, R., Roulet, N., Torn, M.,
635 and Verma, S.: NACP Site: Tower Meteorology, Flux Observations with Uncertainty,
636 and Ancillary Data, Oak Ridge National Laboratory Distributed Active Archive
637 Center, Oak Ridge, Tennessee, USA, 2013.
- 638 Bell, N., Koch, D., and Shindell, D. T.: Impacts of chemistry-aerosol coupling on
639 tropospheric ozone and sulfate simulations in a general circulation model, *J. Geophys.*
640 *Res.*, 110, D14305, doi:10.1029/2004jd005538, 2005.
- 641 Bernacchi, C. J., Kimball, B. A., Quarles, D. R., Long, S. P., and Ort, D. R.: Decreases in
642 stomatal conductance of soybean under open-air elevation of [CO(2)] are closely
643 coupled with decreases in ecosystem evapotranspiration, *Plant Physiol.*, 143, 134-144,
644 doi:10.1104/Pp.106.089557, 2007.
- 645 Bloomer, B. J., Vinnikov, K. Y., and Dickerson, R. R.: Changes in seasonal and diurnal
646 cycles of ozone and temperature in the eastern US, *Atmos. Environ.*, 44, 2543-2551,
647 doi:10.1016/J.Atmosenv.2010.04.031, 2010.
- 648 Bonan, G. B., Lawrence, P. J., Oleson, K. W., Levis, S., Jung, M., Reichstein, M.,
649 Lawrence, D. M., and Swenson, S. C.: Improving canopy processes in the Community
650 Land Model version 4 (CLM4) using global flux fields empirically inferred from
651 FLUXNET data, *J. Geophys. Res.*, 116, G02014, doi:10.1029/2010jg001593, 2011.

652 Chen, M., Zhuang, Q., Cook, D. R., Coulter, R., Pekour, M., Scott, R. L., Munger, J. W.,
653 and Bible, K.: Quantification of terrestrial ecosystem carbon dynamics in the
654 conterminous United States combining a process-based biogeochemical model and
655 MODIS and AmeriFlux data, *Biogeosciences*, 8, 2665-2688, doi:10.5194/Bg-8-2665-
656 2011, 2011.

657 Chiariello, N. R.: Phenology of California grasslands, in: *Grassland structure and*
658 *function: California annual grassland*, edited by: Huenneke, L. F., and Mooney, H. A.,
659 Kluwer Academic Publishers, Dordrecht, 47-58, 1989.

660 Collatz, G. J., Ball, J. T., Grivet, C., and Berry, J. A.: Physiological and Environmental-
661 Regulation of Stomatal Conductance, Photosynthesis and Transpiration - a Model That
662 Includes a Laminar Boundary-Layer, *Agr. Forest Meteorol.*, 54, 107-136,
663 doi:10.1016/0168-1923(91)90002-8, 1991.

664 Cooper, O. R., Parrish, D. D., Stohl, A., Trainer, M., Nedelec, P., Thouret, V., Cammas,
665 J. P., Oltmans, S. J., Johnson, B. J., Tarasick, D., Leblanc, T., McDermid, I. S., Jaffe,
666 D., Gao, R., Stith, J., Ryerson, T., Aikin, K., Campos, T., Weinheimer, A., and Avery,
667 M. A.: Increasing springtime ozone mixing ratios in the free troposphere over western
668 North America, *Nature*, 463, 344-348, doi:10.1038/Nature08708, 2010.

669 Desai, A. R., Noormets, A., Bolstad, P. V., Chen, J. Q., Cook, B. D., Davis, K. J.,
670 Euskirchen, E. S., Gough, C. M., Martin, J. G., Ricciuto, D. M., Schmid, H. P., Tang,
671 J. W., and Wang, W. G.: Influence of vegetation and seasonal forcing on carbon
672 dioxide fluxes across the Upper Midwest, USA: Implications for regional scaling, *Agr.*
673 *Forest Meteorol.*, 148, 288-308, doi:10.1016/J.Agrformet.2007.08.001, 2008.

674 Farquhar, G. D., Caemmerer, S. V., and Berry, J. A.: A Biochemical-Model of
675 Photosynthetic Co₂ Assimilation in Leaves of C-3 Species, *Planta*, 149, 78-90,
676 doi:10.1007/Bf00386231, 1980.

677 Felzer, B., Kicklighter, D., Melillo, J., Wang, C., Zhuang, Q., and Prinn, R.: Effects of
678 ozone on net primary production and carbon sequestration in the conterminous United
679 States using a biogeochemistry model, *Tellus B*, 56, 230-248, doi:10.1111/J.1600-
680 0889.2004.00097.X, 2004.

681 Felzer, B., Reilly, J., Melillo, J., Kicklighter, D., Sarofim, M., Wang, C., Prinn, R., and
682 Zhuang, Q.: Future effects of ozone on carbon sequestration and climate change policy

683 using a global biogeochemical model, *Climatic Change*, 73, 345-373,
684 doi:10.1007/S10584-005-6776-4, 2005.

685 Feng, Z. Z., Kobayashi, K., and Ainsworth, E. A.: Impact of elevated ozone
686 concentration on growth, physiology, and yield of wheat (*Triticum aestivum* L.): a
687 meta-analysis, *Global Change Biol.*, 14, 2696-2708, doi:10.1111/J.1365-
688 2486.2008.01673.X, 2008.

689 Foot, J. P., Caporn, S. J. M., Lee, J. A., and Ashenden, T. W.: The effect of long-term
690 ozone fumigation on the growth, physiology and frost sensitivity of *Calluna vulgaris*,
691 *New Phytol.*, 133, 503-511, doi:10.1111/J.1469-8137.1996.Tb01918.X, 1996.

692 Friend, A. D., and Kiang, N. Y.: Land surface model development for the GISS GCM:
693 Effects of improved canopy physiology on simulated climate, *J Climate*, 18, 2883-
694 2902, doi:10.1175/Jcli3425.1, 2005.

695 Goodale, C. L., Apps, M. J., Birdsey, R. A., Field, C. B., Heath, L. S., Houghton, R. A.,
696 Jenkins, J. C., Kohlmaier, G. H., Kurz, W., Liu, S. R., Nabuurs, G. J., Nilsson, S., and
697 Shvidenko, A. Z.: Forest carbon sinks in the Northern Hemisphere, *Ecol. Appl.*, 12,
698 891-899, Doi 10.2307/3060997, 2002.

699 Grantz, D. A., Vu, H. B., Tew, T. L., and Veremis, J. C.: Sensitivity of Gas Exchange
700 Parameters to Ozone in Diverse C-4 Sugarcane Hybrids, *Crop Sci.*, 52, 1270-1280,
701 doi:10.2135/Cropsci2011.08.0413, 2012.

702 Gregg, J. W., Jones, C. G., and Dawson, T. E.: Urbanization effects on tree growth in the
703 vicinity of New York City, *Nature*, 424, 183-187, doi:10.1038/Nature01728, 2003.

704 Hall, F. G., de Colstoun, E. B., Collatz, G. J., Landis, D., Dirmeyer, P., Betts, A.,
705 Huffman, G. J., Bounoua, L., and Meeson, B.: ISLSCP Initiative II global data sets:
706 Surface boundary conditions and atmospheric forcings for land-atmosphere studies, *J.*
707 *Geophys. Res.*, 111, D22s01, doi:10.1029/2006jd007366, 2006.

708 Heagle, A. S., Kress, L. W., Temple, P. J., Kohut, R. J., Miller, J. E., and Heggestad, H.
709 E.: Factors influencing ozone dose-yield response relationships in open-top chamber
710 studies, in: *Assessment of crop loss from air pollutants*, edited by: Heck, W. W.,
711 Taylor, O. C., and Tingey, D. T., Elsevier Applied Science, New York, 141-179, 1989.

712 Henebry, G. M.: Grasslands of the North American Great Plains, in: Phenology: an
713 integrative environmental science, edited by: Schwartz, M. D., Kluwer Academic
714 Publishers, Dordrecht, 157-174, 2003.

715 Hollaway, M. J., Arnold, S. R., Challinor, A. J., and Emberson, L. D.: Intercontinental
716 trans-boundary contributions to ozone-induced crop yield losses in the Northern
717 Hemisphere, *Biogeosciences*, 9, 271-292, doi:10.5194/Bg-9-271-2012, 2012.

718 Huntingford, C., Cox, P. M., Mercado, L. M., Sitch, S., Bellouin, N., Boucher, O., and
719 Gedney, N.: Highly contrasting effects of different climate forcing agents on terrestrial
720 ecosystem services, *Philos. T. R. Soc. A*, 369, 2026-2037,
721 doi:10.1098/Rsta.2010.0314, 2011.

722 Huntzinger, D. N., Post, W. M., Wei, Y., Michalak, A. M., West, T. O., Jacobson, A. R.,
723 Baker, I. T., Chen, J. M., Davis, K. J., Hayes, D. J., Hoffman, F. M., Jain, A. K., Liu,
724 S., McGuire, A. D., Neilson, R. P., Potter, C., Poulter, B., Price, D., Raczka, B. M.,
725 Tian, H. Q., Thornton, P., Tomelleri, E., Viovy, N., Xiao, J., Yuan, W., Zeng, N.,
726 Zhao, M., and Cook, R.: North American Carbon Program (NACP) regional interim
727 synthesis: Terrestrial biospheric model intercomparison, *Ecol. Model*, 232, 144-157,
728 doi: 10.1016/J.Ecolmodel.2012.02.004, 2012.

729 Ishii, S., Marshall, F. M., and Bell, J. N. B.: Physiological and morphological responses
730 of locally grown Malaysian rice cultivars (*Oryza sativa* L.) to different ozone
731 concentrations, *Water Air Soil Poll.*, 155, 205-221,
732 doi:10.1023/B:Wate.0000026528.86641.5b, 2004.

733 Jaffe, D., and Ray, J.: Increase in surface ozone at rural sites in the western US, *Atmos.*
734 *Environ.*, 41, 5452-5463, doi:10.1016/J.Atmosenv.2007.02.34, 2007.

735 Karnosky, D. F., Skelly, J. M., Percy, K. E., and Chappelka, A. H.: Perspectives
736 regarding 50 years of research on effects of tropospheric ozone air pollution on US
737 forests, *Environ. Pollut.*, 147, 489-506, doi:10.1016/J.Envpol.2006.08.043, 2007.

738 Kvalevag, M. M., and Myhre, G.: The effect of carbon-nitrogen coupling on the reduced
739 land carbon sink caused by tropospheric ozone, *Geophys Res. Lett.*, 40, 3227-3231,
740 doi:10.1002/Grl.50572, 2013.

741 King, A. W., Hayes, D. J., Huntzinger, D. N., West, T. O., and Post, W. M.: North
742 American carbon dioxide sources and sinks: magnitude, attribution, and uncertainty,
743 *Front. Ecol. Environ.*, 10, 512-519, doi: 10.1890/120066, 2012.

744 Knyazikhin, Y., Martonchik, J. V., Myneni, R. B., Diner, D. J., and Running, S. W.:
745 Synergistic algorithm for estimating vegetation canopy leaf area index and fraction of
746 absorbed photosynthetically active radiation from MODIS and MISR data, *J. Geophys.*
747 *Res.*, 103, 32257-32275, doi:10.1029/98jd02462, 1998.

748 Lefohn, A. S., Shadwick, D., and Oltmans, S. J.: Characterizing changes in surface ozone
749 levels in metropolitan and rural areas in the United States for 1980-2008 and 1994-
750 2008, *Atmos. Environ.*, 44, 5199-5210, doi:10.1016/J.Atmosenv.2010.08.049, 2010.

751 Leibensperger, E. M., Mickley, L. J., and Jacob, D. J.: Sensitivity of US air quality to
752 mid-latitude cyclone frequency and implications of 1980-2006 climate change, *Atmos.*
753 *Chem. Phys.*, 8, 7075-7086, 2008.

754 Levy, H., Schwarzkopf, M. D., Horowitz, L., Ramaswamy, V., and Findell, K. L.: Strong
755 sensitivity of late 21st century climate to projected changes in short-lived air
756 pollutants, *J. Geophys. Res.*, 113, D06102, doi:10.1029/2007jd009176, 2008.

757 Lombardozzi, D., Levis, S., Bonan, G., and Sparks, J. P.: Predicting photosynthesis and
758 transpiration responses to ozone: decoupling modeled photosynthesis and stomatal
759 conductance, *Biogeosciences*, 9, 3113-3130, doi:10.5194/bg-9-3113-2012, 2012a.

760 Lombardozzi, D., Sparks, J. P., Bonan, G., and Levis, S.: Ozone exposure causes a
761 decoupling of conductance and photosynthesis: implications for the Ball-Berry
762 stomatal conductance model, *Oecologia*, 169, 651-659, doi:10.1007/S00442-011-
763 2242-3, 2012b.

764 Lombardozzi, D., Sparks, J. P., and Bonan, G.: Integrating O₃ influences on terrestrial
765 processes: photosynthetic and stomatal response data available for regional and global
766 modeling, *Biogeosciences*, 10, 6815-6831, 10.5194/bg-10-6815-2013, 2013.

767 Mulchi, C. L., Slaughter, L., Saleem, M., Lee, E. H., Pausch, R., and Rowland, R.:
768 Growth and Physiological-Characteristics of Soybean in Open-Top Chambers in
769 Response to Ozone and Increased Atmospheric Co₂, *Agr Ecosyst Environ*, 38, 107-
770 118, doi:10.1016/0167-8809(92)90172-8, 1992.

771 Oleson, K. W., Lawrence, D. M., Bonan, G. B., Flanne, M. G., Kluzek, E., Lawrence, P.
772 J., Levis, S., Swenson, S. C., and Thornton, P. E.: Technical Description of version 4.0
773 of the Community Land Model (CLM), National Center for Atmospheric Research,
774 Boulder, CONCAR/TN-478+STR, 2010.

775 Ollinger, S. V., Aber, J. D., Reich, P. B., and Freuder, R. J.: Interactive effects of
776 nitrogen deposition, tropospheric ozone, elevated CO₂ and land use history on the
777 carbon dynamics of northern hardwood forests, *Global Change Biol.*, 8, 545-562,
778 doi:10.1046/J.1365-2486.2002.00482.X, 2002.

779 Pacala, S. W., Hurtt, G. C., Baker, D., Peylin, P., Houghton, R. A., Birdsey, R. A., Heath,
780 L., Sundquist, E. T., Stallard, R. F., Ciais, P., Moorcroft, P., Caspersen, J. P.,
781 Shevliakova, E., Moore, B., Kohlmaier, G., Holland, E., Gloor, M., Harmon, M. E.,
782 Fan, S. M., Sarmiento, J. L., Goodale, C. L., Schimel, D., and Field, C. B.: Consistent
783 land- and atmosphere-based US carbon sink estimates, *Science*, 292, 2316-2320,
784 doi:10.1126/Science.1057320, 2001.

785 Pan, Y. D., Birdsey, R. A., Fang, J. Y., Houghton, R., Kauppi, P. E., Kurz, W. A.,
786 Phillips, O. L., Shvidenko, A., Lewis, S. L., Canadell, J. G., Ciais, P., Jackson, R. B.,
787 Pacala, S. W., McGuire, A. D., Piao, S. L., Rautiainen, A., Sitch, S., and Hayes, D.: A
788 Large and Persistent Carbon Sink in the World's Forests, *Science*, 333, 988-993,
789 doi:10.1126/Science.1201609, 2011.

790 Puma, M. J., Koster, R. D., and Cook, B. I.: Phenological versus meteorological controls
791 on land-atmosphere water and carbon fluxes, *J. Geophys. Res.*, 118, 14-29,
792 doi:10.1029/2012jg002088, 2013.

793 Reichle, R. H., Koster, R. D., De Lannoy, G. J. M., Forman, B. A., Liu, Q., Mahanama,
794 S. P. P., and Toure, A.: Assessment and Enhancement of MERRA Land Surface
795 Hydrology Estimates, *J Climate*, 24, 6322-6338, doi:10.1175/Jcli-D-10-05033.1, 2011.

796 Ricciuto, D., Schaefer, K., Thornton, P., Davis, K., Cook, R., Liu, S., Anderson, R.,
797 Arain, M., Baker, I., Chen, J., Dietze, M., Grant, R., Izaurrealde, C., Jain, A., King, A.,
798 Kucharik, C., Liu, S., Lokupitiya, E., Luo, Y., Peng, C., Poulter, B., Price, D., Riley,
799 W., Sahoo, A., Tian, H., Tonitto, C., and Verbeeck, H.: NACP Site: Terrestrial
800 Biosphere Model and Aggregated Flux Data in Standard Format, Oak Ridge National
801 Laboratory Distributed Active Archive Center, Oak Ridge, Tennessee, USA, 2013.

802 Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E.,
803 Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G. K., Bloom, S., Chen, J. Y.,
804 Collins, D., Conaty, A., Da Silva, A., Gu, W., Joiner, J., Koster, R. D., Lucchesi, R.,
805 Molod, A., Owens, T., Pawson, S., Pegion, P., Redder, C. R., Reichle, R., Robertson,
806 F. R., Ruddick, A. G., Sienkiewicz, M., and Woollen, J.: MERRA: NASA's Modern-
807 Era Retrospective Analysis for Research and Applications, *J Climate*, 24, 3624-3648,
808 doi:10.1175/Jcli-D-11-00015.1, 2011.

809 Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J.,
810 Cunnold, D. M., Steele, L. P., Krummel, P. B., Weiss, R. F., O'Doherty, S., Salameh,
811 P. K., Wang, H. J., Harth, C. M., Muhle, J., and Porter, L. W.: Renewed growth of
812 atmospheric methane, *Geophys. Res. Lett.*, 35, L22805, doi:10.1029/2008gl036037,
813 2008.

814 Rudorff, B. F. T., Mulchi, C. L., Lee, E. H., Rowland, R., and Pausch, R.: Effects of
815 enhanced O₃ and CO₂ enrichment on plant characteristics in wheat and corn,
816 *Environ. Pollut.*, 94, 53-60, doi:10.1016/S0269-7491(96)00050-4, 1996.

817 Schaefer, K., Schwalm, C. R., Williams, C., Arain, M. A., Barr, A., Chen, J. M., Davis,
818 K. J., Dimitrov, D., Hilton, T. W., Hollinger, D. Y., Humphreys, E., Poulter, B.,
819 Raczka, B. M., Richardson, A. D., Sahoo, A., Thornton, P., Vargas, R., Verbeeck, H.,
820 Anderson, R., Baker, I., Black, T. A., Bolstad, P., Chen, J. Q., Curtis, P. S., Desai, A.
821 R., Dietze, M., Dragoni, D., Gough, C., Grant, R. F., Gu, L. H., Jain, A., Kucharik, C.,
822 Law, B., Liu, S. G., Lokipitiya, E., Margolis, H. A., Matamala, R., McCaughey, J. H.,
823 Monson, R., Munger, J. W., Oechel, W., Peng, C. H., Price, D. T., Ricciuto, D., Riley,
824 W. J., Roulet, N., Tian, H. Q., Tonitto, C., Torn, M., Weng, E. S., and Zhou, X. L.: A
825 model-data comparison of gross primary productivity: Results from the North
826 American Carbon Program site synthesis, *J. Geophys. Res.*, 117, G03010,
827 doi:10.1029/2012jg001960, 2012.

828 Shindell, D. T., Faluvegi, G., Unger, N., Aguilar, E., Schmidt, G. A., Koch, D. M., Bauer,
829 S. E., and Miller, R. L.: Simulations of preindustrial, present-day, and 2100 conditions
830 in the NASA GISS composition and climate model G-PUCCINI, *Atmos. Chem. Phys.*,
831 6, 4427-4459, 2006.

832 Shindell, D. T., Pechony, O., Voulgarakis, A., Faluvegi, G., Nazarenko, L., Lamarque, J.
833 F., Bowman, K., Milly, G., Kovari, B., Ruedy, R., and Schmidt, G. A.: Interactive
834 ozone and methane chemistry in GISS-E2 historical and future climate simulations,
835 *Atmos. Chem. Phys.*, 13, 2653-2689, doi:10.5194/Acp-13-2653-2013, 2013.

836 Sitch, S., Cox, P. M., Collins, W. J., and Huntingford, C.: Indirect radiative forcing of
837 climate change through ozone effects on the land-carbon sink, *Nature*, 448, 791-794,
838 doi:10.1038/Nature06059, 2007.

839 Spitters, C. J. T., Toussaint, H. A. J. M., and Goudriaan, J.: Separating the Diffuse and
840 Direct Component of Global Radiation and Its Implications for Modeling Canopy
841 Photosynthesis .1. Components of Incoming Radiation, *Agr. Forest Meteorol.*, 38,
842 217-229, doi:10.1016/0168-1923(86)90060-2, 1986.

843 Taylor, M. D., Sinn, J. P., Davis, D. D., and Pell, E. J.: The impact of ozone on a salt
844 marsh cordgrass (*Spartina alterniflora*), *Environ. Pollut.*, 120, 701-705, 2002.

845 Unger, N.: Global climate impact of civil aviation for standard and desulfurized jet fuel,
846 *Geophys. Res. Lett.*, 38, L20803, doi:10.1029/2011gl049289, 2011.

847 Unger, N., and Pan, J. L.: New Directions: Enduring ozone, *Atmos. Environ.*, 55, 456-
848 458, doi:10.1016/J.Atmosenv.2012.03.036, 2012.

849 Unger, N., Harper, K., Zheng, Y., Kiang, N. Y., Aleinov, I., Arneth, A., Schurgers, G.,
850 Amelynck, C., Goldstein, A., Guenther, A., Heinesch, B., Hewitt, C. N., Karl, T.,
851 Laffineur, Q., Langford, B., McKinney, K. A., Misztal, P., Potosnak, M., Rinne, J.,
852 Pressley, S., Schoon, N., and Serça, D.: Photosynthesis-dependent isoprene emission
853 from leaf to planet in a global carbon–chemistry–climate model, *Atmos. Chem. Phys.*,
854 13, 10243-10269, doi:10.5194/acp-13-10243-2013, 2013.

855 von Caemmerer, S., and Farquhar, G. D.: Some Relationships between the Biochemistry
856 of Photosynthesis and the Gas-Exchange of Leaves, *Planta*, 153, 376-387, 1981.

857 VanLoocke, A., Betzelberger, A. M., Ainsworth, E. A., and Bernacchi, C. J.: Rising
858 ozone concentrations decrease soybean evapotranspiration and water use efficiency
859 whilst increasing canopy temperature, *New Phytol.*, 195, 164-171, doi:10.1111/J.1469-
860 8137.2012.04152.X, 2012.

861 Wittig, V. E., Ainsworth, E. A., and Long, S. P.: To what extent do current and projected
862 increases in surface ozone affect photosynthesis and stomatal conductance of trees? A

863 meta-analytic review of the last 3 decades of experiments, *Plant Cell Environ.*, 30,
864 1150-1162, doi:10.1111/J.1365-3040.2007.01717.X, 2007.

865 Wu, S. L., Mickley, L. J., Leibensperger, E. M., Jacob, D. J., Rind, D., and Streets, D. G.:
866 Effects of 2000-2050 global change on ozone air quality in the United States, *J.*
867 *Geophys. Res.*, 113, D06302, doi:10.1029/2007jd008917, 2008.

868 Xiao, J. F., Zhuang, Q. L., Law, B. E., Chen, J. Q., Baldocchi, D. D., Cook, D. R., Oren,
869 R., Richardson, A. D., Wharton, S., Ma, S. Y., Martin, T. A., Verma, S. B., Suyker, A.
870 E., Scott, R. L., Monson, R. K., Litvak, M., Hollinger, D. Y., Sun, G., Davis, K. J.,
871 Bolstad, P. V., Burns, S. P., Curtis, P. S., Drake, B. G., Falk, M., Fischer, M. L.,
872 Foster, D. R., Gu, L. H., Hadley, J. L., Katul, G. G., Roser, Y., McNulty, S., Meyers,
873 T. P., Munger, J. W., Noormets, A., Oechel, W. C., Paw, K. T., Schmid, H. P., Starr,
874 G., Torn, M. S., and Wofsy, S. C.: A continuous measure of gross primary production
875 for the conterminous United States derived from MODIS and AmeriFlux data, *Remote*
876 *Sens. Environ.*, 114, 576-591, doi:10.1016/J.Rse.2009.10.013, 2010.

877 Zhang, L., Su, B. Y., Xu, H., and Li, Y. G.: Growth and photosynthetic responses of four
878 landscape shrub species to elevated ozone, *Photosynthetica*, 50, 67-76,
879 doi:10.1007/S11099-012-0004-Z, 2012.

880
881

882

883

884

885 **Table 1.** Parameters for vegetation model and O₃ damage scheme

886

| PFT ^a | TDA | GRAC3 | GRAC4 | SHR | DBF | ENF | TRF | CRO | |
|---|------|-------|-------|------|------|-------|------|-----------------|-----------------|
| Carboxylation | C3 | C3 | C4 | C3 | C3 | C3 | C3 | C3 ^b | C4 ^b |
| V_{max25} ($\mu\text{mol m}^{-2} \text{s}^{-1}$) | 33 | 43 | 24 | 25 | 30 | 43 | 75 | 40 | 40 |
| m | 9 | 11 | 5 | 9 | 9 | 9 | 9 | 11 | 5 |
| b ($\text{mmol m}^{-2} \text{s}^{-1}$) | 2 | 8 | 2 | 2 | 2 | 2 | 2 | 8 | 2 |
| O3T ($\text{nmol m}^{-2} \text{s}^{-1}$) | 1.6 | 5 | 5 | 1.6 | 1.6 | 1.6 | 1.6 | 5 | 5 |
| a (high) ($\text{mmol}^{-1} \text{m}^{-2}$) | 0.1 | 1.4 | 0.735 | 0.1 | 0.15 | 0.075 | 0.15 | 1.4 | 0.735 |
| a (low) ($\text{mmol}^{-1} \text{m}^{-2}$) | 0.03 | 0.25 | 0.13 | 0.03 | 0.04 | 0.02 | 0.04 | 0.25 | 0.13 |

887

888 ^a Plant function types (PFTs) are tundra (TDA), C3 grassland (GRAC3), C4
889 savanna/grassland (GRAC4), shrubland (SHR), deciduous broadleaf forest (DBF),
890 evergreen needleleaf forest (ENF), tropical rainforest (TRF), and cropland (CRO).

891 ^b For site-level simulations, we consider CRO as C4 plant. For regional simulation, we
892 consider half CRO as C3 plants (soybean) and the rest C4 plant (corn).

893

894

895

896
 897
 898
 899
 900
 901

Table 2. Description of the site-level simulations.

| ID | Simulations ^a | Meteorology | | Vegetation (LAI) | | |
|----|-----------------------------------|-------------|-------|------------------|-------|-----------------------------------|
| | | Site | MERRA | MODIS | MERRA | Incl. O ₃ ^c |
| 1 | METmerra_LAImodis | | Yes | Yes | | |
| 2 | METsite_LAImodis ^b | Yes | Yes | Yes | | |
| 3 | METmerra_LAIterra | | Yes | | Yes | |
| 4 | METsite_LAIterra ^b | Yes | Yes | | Yes | |
| 5 | METsite_LAIterra_LO3 ^b | Yes | Yes | | Yes | Low ^d |
| 6 | METsite_LAIterra_HO3 ^b | Yes | Yes | | Yes | High ^d |

902

903 ^a The name of each simulation is composed of at least two words. The prefix indicates the
 904 source of meteorological forcings. The suffix or the second word indicates the sources of
 905 vegetation forcings.

906 ^b For simulations with prefix ‘SITE’ use site-based meteorological forcings, which are
 907 gap-filled with MERRA-land reanalyses.

908 ^c Ambient [O₃] is applied at each site.

909 ^d Low and high indicate the sensitivity of GPP to [O₃] defined by the coefficient *a* in
 910 Table 1.

911

912

913

914

915

916 **Table 3.** Statistics ^a for site-level simulations.

917

| ID | Simulations | R ² | | | RMSE | | | χ ² | | |
|----|----------------------|----------------|------|------|------|-----|------|----------------|------|-------|
| | | min | max | mean | min | max | mean | min | max | total |
| 1 | METmerra_LAImodis | 0.14 | 0.85 | 0.63 | 1.2 | 6.4 | 3.0 | 1.4 | 37.6 | 9.4 |
| 2 | METsite_LAImodis | 0.16 | 0.85 | 0.67 | 1.1 | 5.8 | 2.9 | 1.1 | 39.6 | 8.9 |
| 3 | METmerra_LAIterra | 0 | 0.88 | 0.66 | 1.3 | 4.2 | 2.4 | 1.2 | 16.6 | 4.8 |
| 4 | METsite_LAIterra | 0 | 0.87 | 0.68 | 1.0 | 4.2 | 2.3 | 1.2 | 13.4 | 4.1 |
| 5 | METsite_LAIterra_LO3 | 0 | 0.88 | 0.69 | 1.0 | 4.1 | 2.3 | 1.1 | 13.0 | 3.9 |
| 6 | METsite_LAIterra_HO3 | 0 | 0.88 | 0.69 | 1.0 | 4.1 | 2.2 | 1.0 | 12.3 | 3.7 |

918

919 ^a Statistics include minimum and maximum values of R², RMSE, and χ² for 35 NCAP
920 sites with χ² < 16 (Fig. 3a). We also calculate the mean values of R² and RMSE for these
921 sites. We calculate the total χ² (shown as red bars in Fig. 4) using all the available
922 observations over all sites.

923

924

925 **Appendix**926 **Table A.** Descriptions of NACP sites in Canada ^a

| Site | PFT ^b | Description | Longitude | Latitude | Period |
|--------|------------------|----------------------------|-----------|----------|-----------|
| CA-Ca1 | ENF | Campbell River | 125.3°W | 49.9°N | 1998-2006 |
| CA-Ca2 | ENF | Campbell River | 125.3°W | 49.9°N | 2001-2006 |
| CA-Ca3 | ENF | Campbell River | 124.9°W | 49.5°N | 2002-2006 |
| CA-Gro | MF | Groundhog River | 82.2°W | 48.2°N | 2004-2006 |
| CA-Let | GRA | Lethbridge Grassland | 112.9°W | 49.7°N | 2001-2007 |
| CA-Mer | WET | Eastern Peatland | 75.5°W | 45.4°N | 1999-2006 |
| CA-NS1 | ENF | UCI Chronosequence | 124.9°W | 49.5°N | 2001-2005 |
| CA-Oas | DBF | BERMS | 106.2°W | 53.6°N | 1997-2006 |
| CA-Obs | ENF | BERMS | 105.1°W | 54.0°N | 2000-2006 |
| CA-Ojp | ENF | BERMS | 104.7°W | 53.9°N | 2000-2006 |
| CA-Qfo | ENF | Quebec | 74.3°W | 49.7°N | 2004-2006 |
| CA-SJ1 | ENF | BERMS | 104.7°W | 53.9°N | 2002-2005 |
| CA-SJ2 | ENF | BERMS | 104.6°W | 53.9°N | 2004-2006 |
| CA-SJ3 | ENF | BERMS | 104.6°W | 53.9°N | 2005-2006 |
| CA-TP4 | ENF | Turkey Point | 80.4°W | 42.7°N | 2003-2007 |
| CA-WP1 | WET | Western Peatland | 112.5°W | 55.0°N | 2004-2007 |
| US-ARM | GRA ^c | Southern Great Plains | 97.5°W | 36.6°N | 2003-2007 |
| US-Dk3 | ENF | Duke Forest | 79.1°W | 36.0°N | 1998-2005 |
| US-Ha1 | DBF | Harvard Forest | 72.2°W | 42.5°N | 1992-2006 |
| US-Ho1 | ENF | Howland Forest | 68.7°W | 45.2°N | 1996-2004 |
| US-IB1 | CRO | Fermi Lab | 88.2°W | 41.9°N | 2006 |
| US-IB2 | GRA | Fermi | 88.2°W | 41.8°N | 2005-2006 |
| US-Los | WET | Lost Creek | 90.0°W | 46.1°N | 2001-2006 |
| US-MMS | DBF | Morgan Monroe State Forest | 86.4°W | 39.3°N | 1999-2006 |
| US-MOz | DBF | Missouri Ozark | 92.2°W | 38.7°N | 2005-2007 |
| US-Me2 | ENF | Metolius | 121.6°W | 44.5°N | 2002-2007 |
| US-Me3 | ENF | Metolius | 121.6°W | 44.3°N | 2004-2005 |
| US-Me5 | ENF | Metolius | 121.6°W | 44.4°N | 2000-2002 |
| US-NR1 | ENF | Niwot Ridge | 105.5°W | 40.0°N | 1999-2007 |
| US-Ne1 | CRO | Mead | 96.5°W | 41.2°N | 2002-2005 |
| US-Ne2 | CRO | Mead | 96.5°W | 41.2°N | 2003-2005 |
| US-Ne3 | CRO | Mead | 96.4°W | 41.2°N | 2002-2005 |
| US-Pfa | MF | Park Falls | 90.3°W | 45.9°N | 1997-2004 |
| US-SO2 | CSH | Sky Oaks | 116.6°W | 33.4°N | 1999-2006 |
| US-Shd | GRA | Shidler | 96.7°W | 36.9°N | 1998-1999 |
| US-Syv | MF | Sylvania Wilderness Area | 89.3°W | 46.2°N | 2002-2006 |
| US-Ton | WSA | Tonzi Ranch | 121.0°W | 38.4°N | 2002-2007 |
| US-UMB | DBF | UMBS | 84.7°W | 45.6°N | 1999-2006 |
| US-Var | GRA | Varia Ranch | 121.0°W | 38.4°N | 2001-2007 |
| US-WCr | DBF | Willow Creek | 90.1°W | 45.8°N | 1999-2006 |

927

928 ^a Site information is adopted from Schaefer et al. (2012), except that the operational time
929 span listed here is only for the period when measurements of GPP are available.930 ^b PFT names are: evergreen needleleaf forest (ENF), deciduous broadleaf forest (DBF),
931 grasslands (GRA), croplands (CRO), closed shrublands (CSH), mixed forests (MF),
932 permanent wetlands (WET), and woody savannas (WSA).

933 ^c The land type at US-ARM is cropland in Schaefer et al. (2012). However, the site is
934 covered by cattle pasture and wheat fields (<https://www.arm.gov/sites/sgp>), which are
935 more like C3 grassland.

936

937

938

939 **Figure Captions**

940 **Fig. 1.** Comparison of monthly mean leaf area index (LAI, $\text{m}^2 \text{m}^{-2}$) from the Modern Era-
941 Retrospective Analysis (MERRA) reanalysis (red solid lines) and the Moderate
942 Resolution Imaging Spectroradiometer (MODIS) (blue dashed lines) at each NACP site.
943 The name and location (longitude, latitude) are shown in the title. The land types include
944 evergreen needleleaf forest (ENF), deciduous broadleaf forest (DBF), shrublands (SHR),
945 grasslands (GRA), and croplands (CRO).

946

947 **Fig. 2.** Comparison of long-term monthly mean GPP ($\text{g [C] m}^{-2} \text{day}^{-1}$) from observations
948 (blue points with error bars indicating one standard deviation) and the O_3 -free simulations
949 (black lines with shadings indicating one standard deviation) averaged over measurement
950 period at each site. The simulation, METsite_LAI_{merra}, is driven with meteorological
951 forcings from MERRA and site measurements. The LAI is from MERRA. The name and
952 location (longitude, latitude) are shown in the title. The land types include evergreen
953 needleleaf forest (ENF), deciduous broadleaf forest (DBF), shrublands (SHR), grasslands
954 (GRA), and croplands (CRO).

955

956 **Fig. 3.** The calculated (a) χ^2 of GPP and changes in χ^2 after the inclusion of O_3 damage
957 impact with (b) low and (c) high O_3 sensitivity at each site. The sites are sorted according
958 to the values of χ^2 in (a). The land cover definitions are: GRA, Grasslands; CRO,
959 Croplands; ENF, Evergreen Needleleaf Forest; DBF, Deciduous Broadleaf Forest; SHR,
960 Shrubland. See section 2.2.1 for the definition of χ^2 .

961

962 **Fig. 4.** The calculated average χ^2 of GPP over NACP sites for 6 different simulations as
963 listed in Table 2. The blue bars are results for all 40 NACP sites. The red bars are results
964 excluding sites CA-Let, CA-NS1, US-Var, CA-SJ1, and CA-SJ2, where the simulated
965 site-level χ^2 is larger than 16 as shown in Fig. 3a.

966

967 **Fig. 5.** Validation of simulated summertime surface (a, b) diurnal mean and (c, d) daily
968 maximum 8-hour average O_3 with *in situ* measurements from (a, b) the EPA Clean Air
969 Status and Trends Network (CASTNET) and (c, d) the AIRDATA. For (b) and (d), the

970 blue points indicate sites East of 95°W and the red ones for West of 95°W. The
971 correlation coefficients between simulations and observations are shown in (b) and (d).
972 Please refer to Fig. S5 for separate results from GCM and observations.

973

974 **Fig. 6.** Simulated summertime (a) surface [O₃], (b) stomatal conductance, (c) ozone
975 stomatal flux, and (d) damages to GPP at different O₃ sensitivity for 24 U.S. sites. The
976 sites are sorted according to the simulated O₃-free GPP in (d). For each site, the result is
977 averaged over the period when the site provides GPP measurements. The land cover
978 definitions are: GRA, Grasslands; CRO, Croplands; ENF, Evergreen Needleleaf Forest;
979 DBF, Deciduous Broadleaf Forest; SHR, Shrubland.

980

981 **Fig. 7.** Changes in GPP for all and individual PFTs in the presence of different levels of
982 [O₃] as simulated by the vegetation model. Simulations are performed at 40 NACP sites
983 with a fixed [O₃] for either low or high O₃ sensitivity. The short blue lines show the
984 damages ranging from low to high O₃ sensitivity, with the blue points indicating the
985 average reductions. The simulation results are averaged for all the sites or for the sites
986 with the same PFT. The number of sites used for average is shown in the title bracket of
987 each subplot. The solid squares with lines show the results (mean plus uncertainty) based
988 on measurements reported by multiple literatures. These measurements are collected by
989 Lombardozzi et al. (2013) for all PFTs, Wittig et al. (2007) for evergreen needleleaf
990 forest (ENF) and deciduous broadleaf forest (DBF). For C4 grass or crop (CRO_C4), we
991 have collected data from Taylor et al. (2002) for spartina alterniflora and Grantz et al.
992 (2012) for sugarcane hybrids. For C3 grass or crop (GRA_C3), we have collected data
993 from Feng et al. (2008) for wheat, Foot et al. (1996) for colluna vulgaris, Mulchi et al.
994 (1992) for soybean, and Ishii et al. (2004) and Ainsworth (2008) for rice. The values for
995 rice are denoted in green and others in red. The author initials are indicated for the
996 corresponding studies.

997

998 **Fig. 8.** Simulated summertime (a) O₃-exposed GPP and (b) O₃ stomatal flux over the U.S.
999 The simulated GPP is overlaid with *in situ* measurements from NACP. The simulations

1000 are performed with land cover from ISLSCP and meteorological forcings from MERRA
1001 reanalyses. Please refer to Fig. S6 for separate results from simulations and observations.

1002

1003 **Fig. 9.** Simulated reduction fraction in summer GPP in the U.S. due to (a) low and (b)
1004 high O₃ sensitivity for 1998-2007.

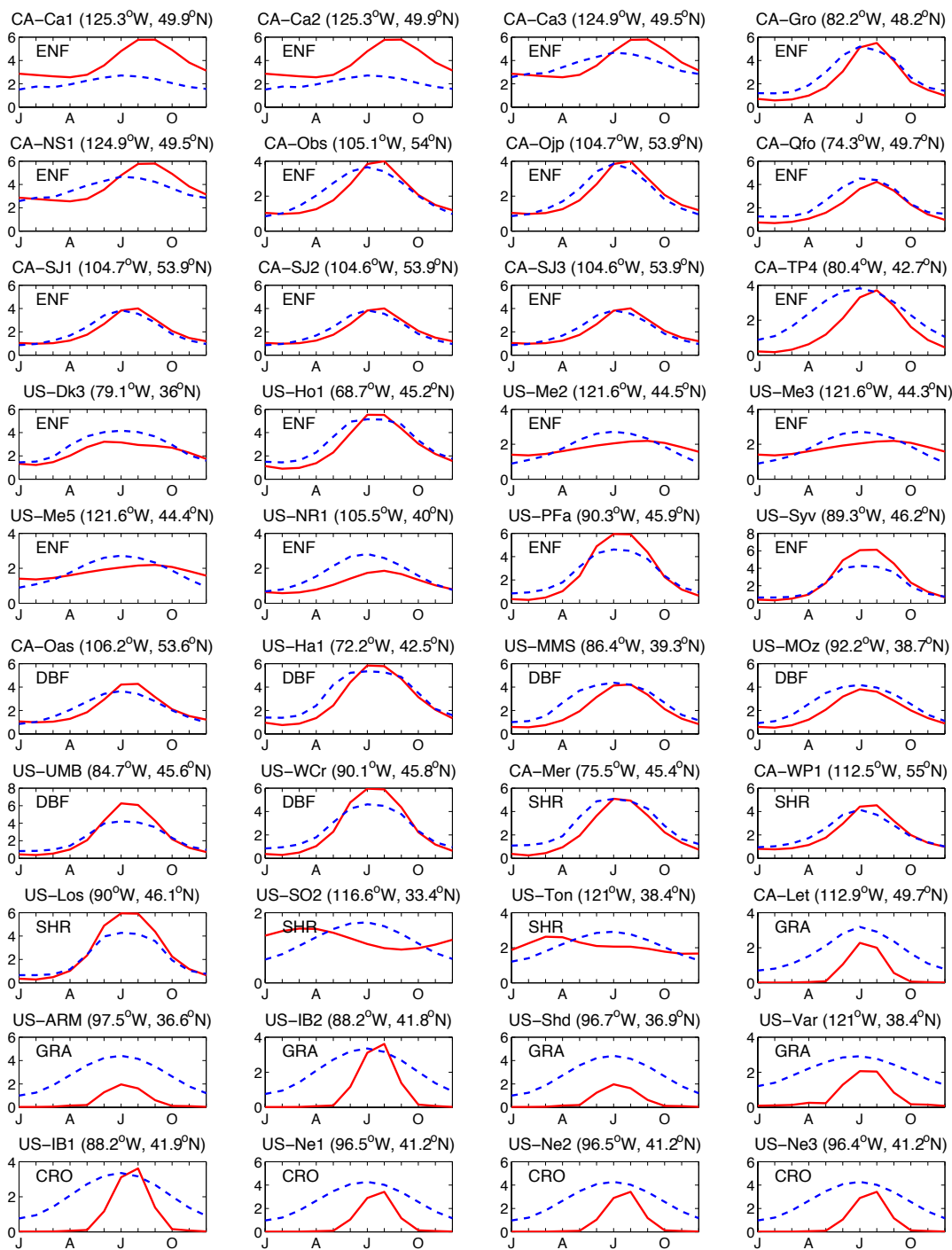
1005

1006 **Fig. 10.** Simulated changes in summer GPP due to (a, c) 25% reduction or (b, d) 25%
1007 increase in [O₃] for (a, b) low or (c, d) high O₃ sensitivity.

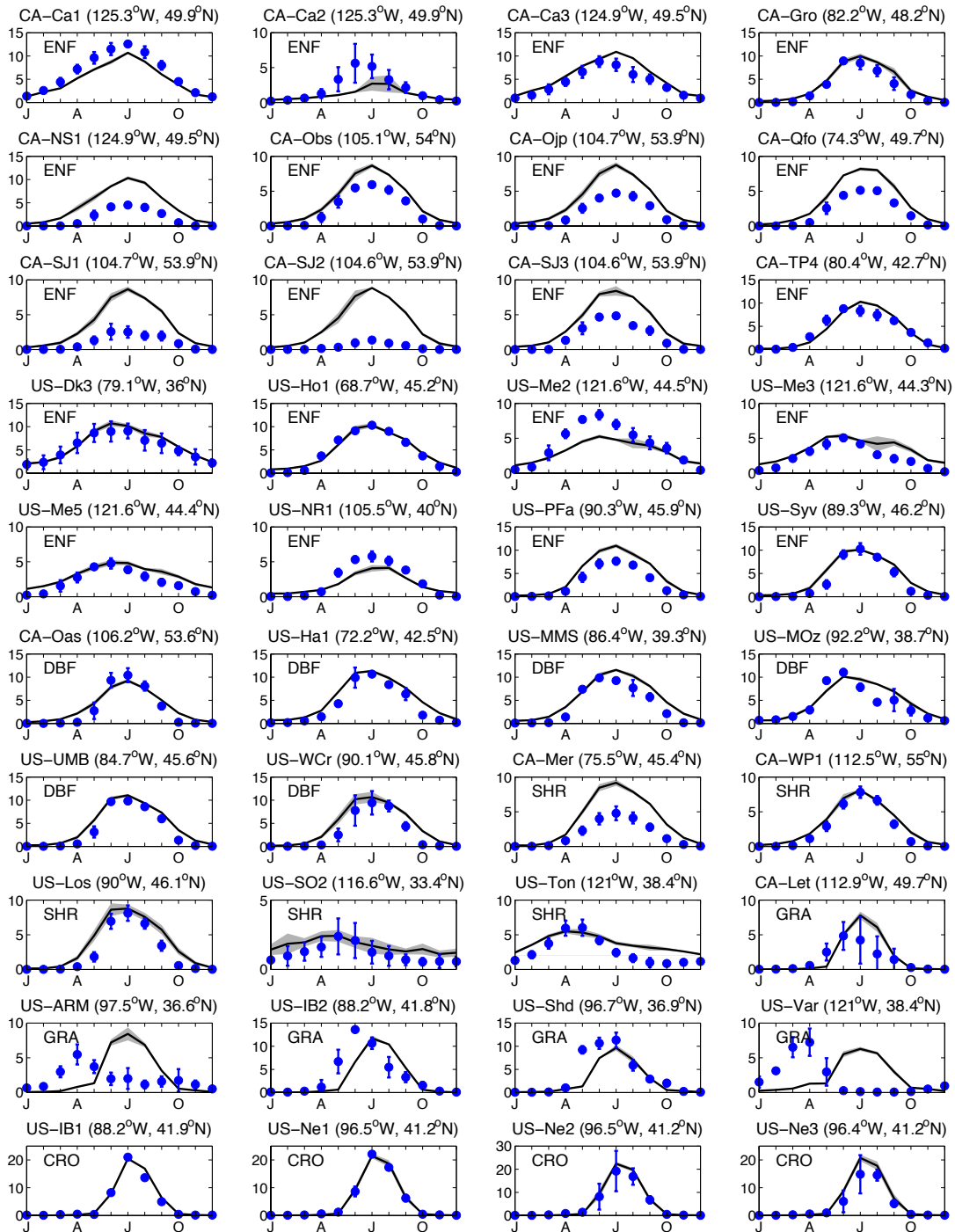
1008

1009

1010
 1011
 1012
 1013
 1014
 1015
 1016
 1017
 1018
 1019
 1020
 1021
 1022
 1023
 1024
 1025
 1026
 1027
 1028
 1029
 1030
 1031
 1032
 1033
 1034
 1035
 1036
 1037
 1038



1. Comparison of monthly mean leaf area index (LAI, $m^2 m^{-2}$) from the Modern Era-retrospective Analysis (MERRA) reanalysis (red solid lines) and the Moderate Resolution Imaging Spectroradiometer (MODIS) (blue dashed lines) at each NACP site. The name and location (longitude, latitude) are shown in the title. The land types include evergreen needleleaf forest (ENF), deciduous broadleaf forest (DBF), shrublands (SHR), grasslands (GRA), and croplands (CRO).



1039 **Fig. 2.** Comparison of long-term monthly mean GPP ($\text{g [C] m}^{-2} \text{ day}^{-1}$) from observations
 1040 (blue points with error bars indicating one standard deviation) and the O_3 -free simulations
 1041 (black lines with shadings indicating one standard deviation) averaged over measurement
 1042 period at each site. The simulation, METsite_LAIterra, is driven with meteorological
 1043 forcings from MERRA and site measurements. The LAI is from MERRA. The name and
 1044 location (longitude, latitude) are shown in the title. The land types include evergreen
 1045 needleleaf forest (ENF), deciduous broadleaf forest (DBF), shrublands (SHR), grasslands
 1046 (GRA), and croplands (CRO).

1047
 1048
 1049
 1050
 1051
 1052
 1053
 1054
 1055
 1056
 1057
 1058
 1059
 1060
 1061
 1062
 1063
 1064
 1065
 1066
 1067
 1068
 1069
 1070
 1071
 1072
 1073
 1074
 1075
 1076
 1077
 1078
 1079
 1080
 1081
 1082
 1083
 1084
 1085
 1086
 1087
 1088
 1089
 1090
 1091
 1092

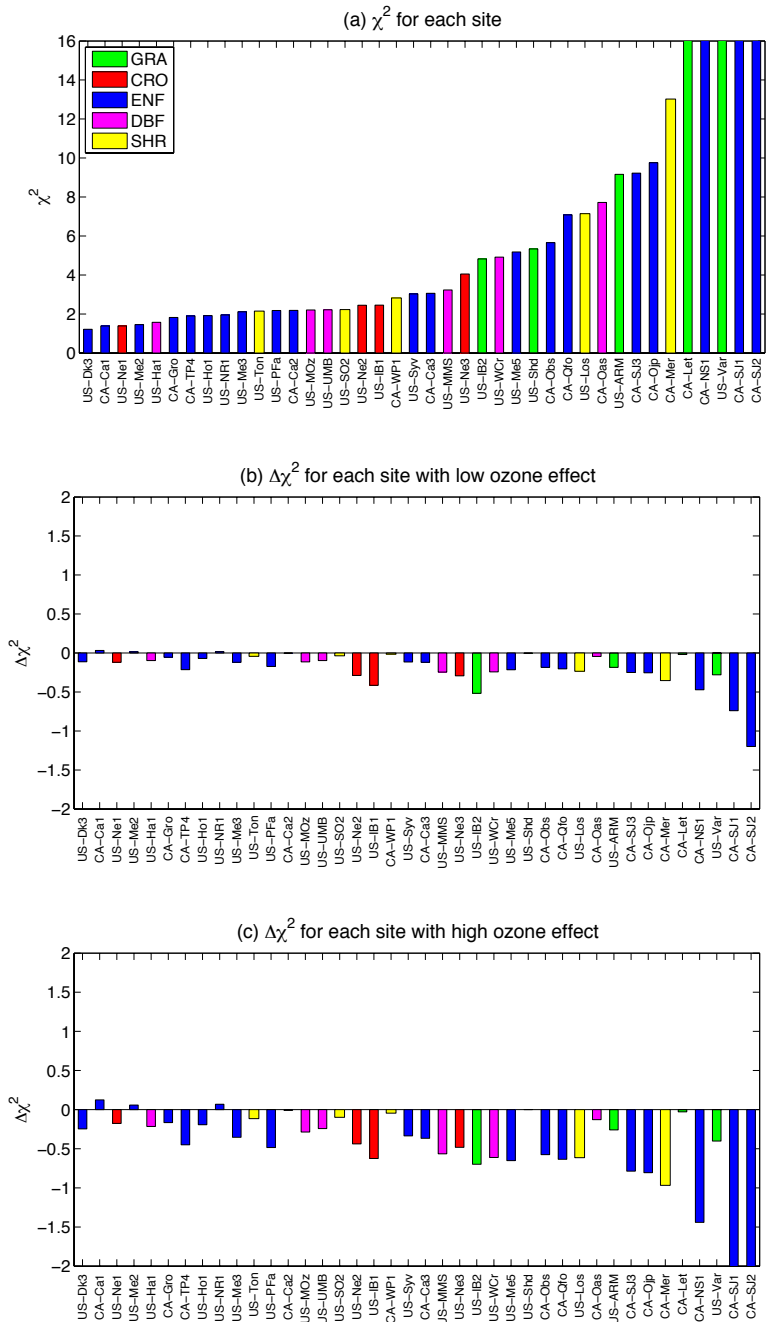


Fig. 3. The calculated (a) χ^2 of GPP and changes in χ^2 after the inclusion of O_3 damage impact with (b) low and (c) high O_3 sensitivity at each site. The sites are sorted according to the values of χ^2 in (a). The land cover definitions are: GRA, Grasslands; CRO, Croplands; ENF, Evergreen Needleleaf Forest; DBF, Deciduous Broadleaf Forest; SHR, Shrubland. See section 2.2.1 for the definition of χ^2 .

1093
 1094
 1095
 1096
 1097
 1098
 1099
 1100
 1101
 1102
 1103
 1104
 1105
 1106
 1107
 1108
 1109
 1110
 1111
 1112
 1113
 1114
 1115
 1116
 1117
 1118

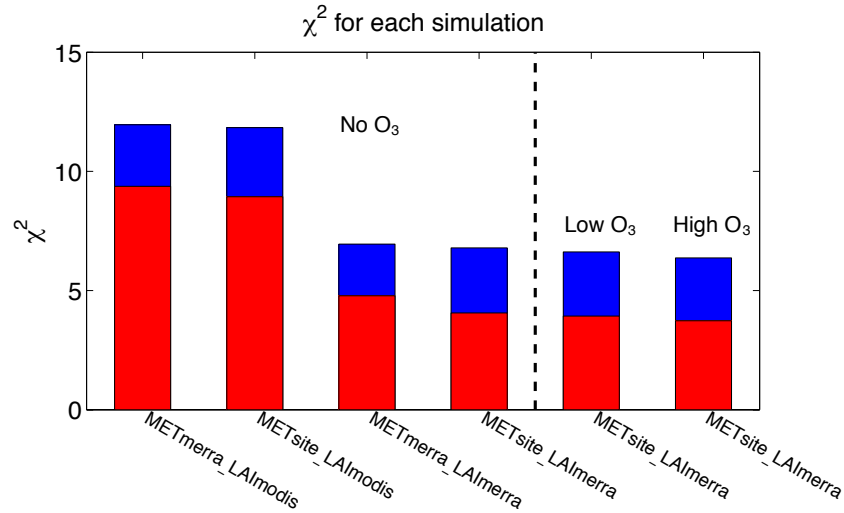


Fig. 4. The calculated average χ^2 of GPP over NACP sites for 6 different simulations as listed in Table 2. The blue bars are results for all 40 NACP sites. The red bars are results excluding sites CA-Let, CA-NS1, US-Var, CA-SJ1, and CA-SJ2, where the simulated site-level χ^2 is larger than 16 as shown in Fig. 3a.

1119
1120
1121
1122
1123
1124
1125
1126
1127
1128
1129
1130
1131
1132
1133
1134
1135
1136
1137
1138
1139
1140
1141
1142
1143
1144
1145
1146
1147
1148
1149
1150
1151
1152

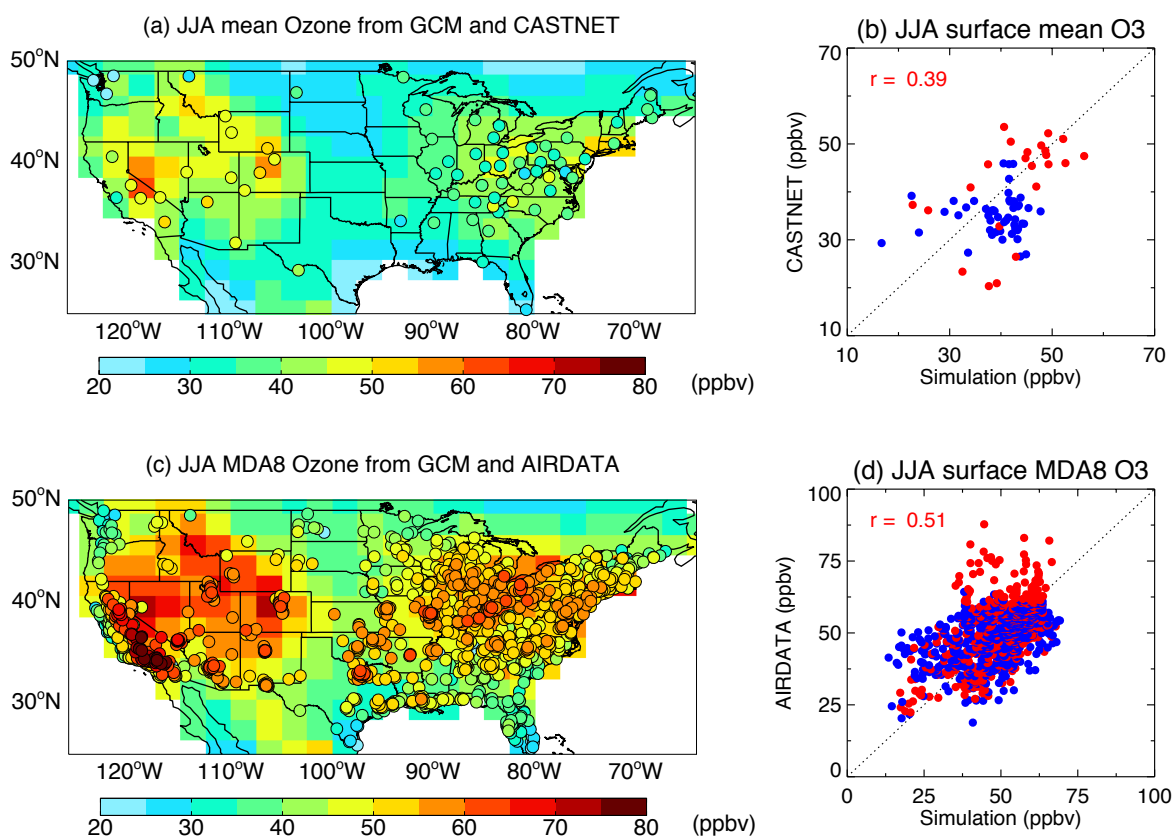
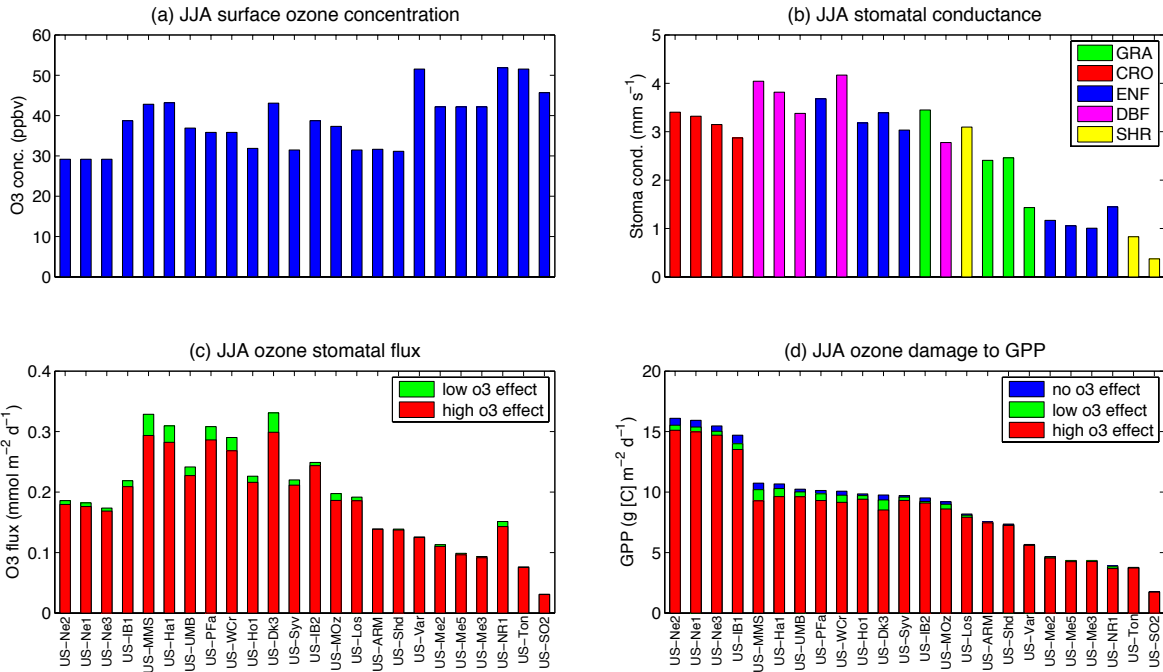


Fig. 5. Validation of simulated summertime surface (a, b) diurnal mean and (c, d) daily maximum 8-hour average O₃ with *in situ* measurements from (a, b) the EPA Clean Air Status and Trends Network (CASTNET) and (c, d) the AIRDATA. For (b) and (d), the blue points indicate sites East of 95°W and the red ones for West of 95°W. The correlation coefficients between simulations and observations are shown in (b) and (d). Please refer to Fig. S5 for separate results from GCM and observations.

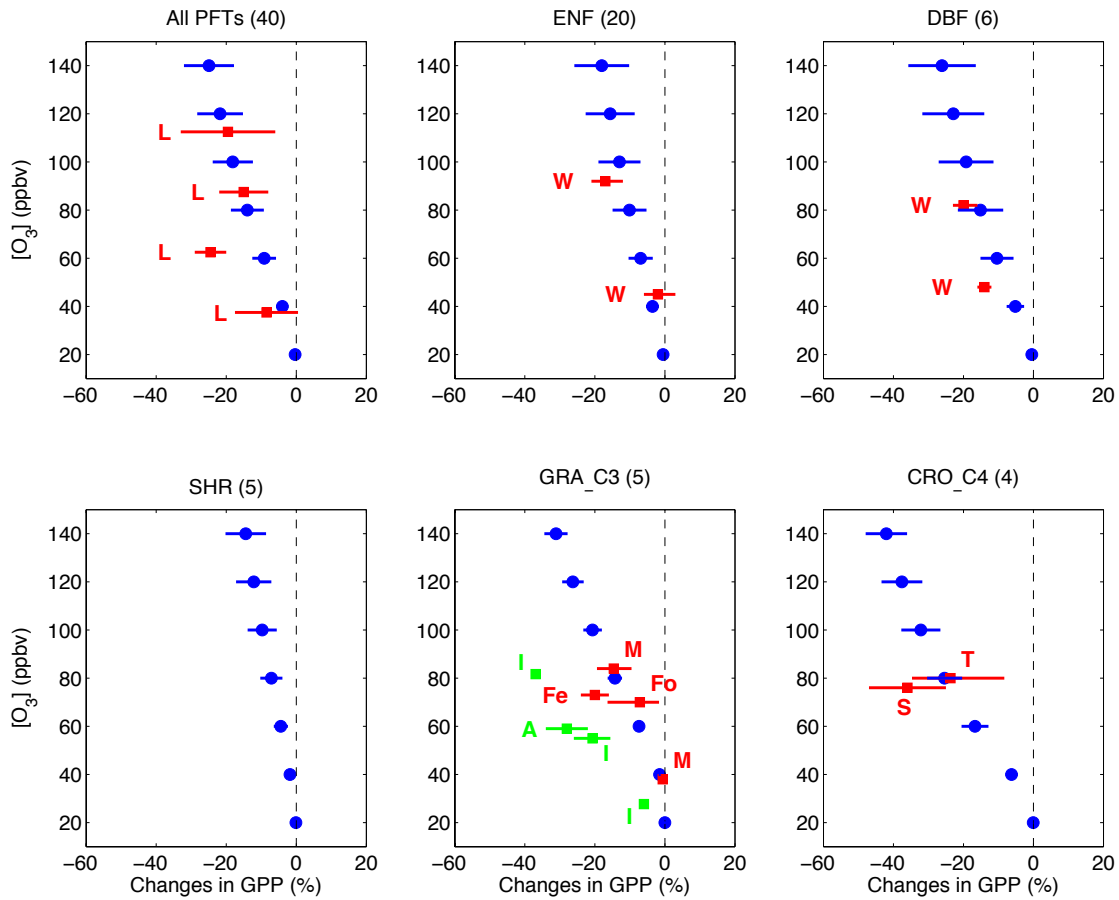
1153
1154
1155



1156

1157
1158
1159
1160
1161
1162
1163
1164
1165
1166

Fig. 6. Simulated summertime (a) surface $[O_3]$, (b) stomatal conductance, (c) ozone stomatal flux, and (d) damages to GPP at different O_3 sensitivity for 24 U.S. sites. The sites are sorted according to the simulated O_3 -free GPP in (d). For each site, the result is averaged over the period when the site provides GPP measurements. The land cover definitions are: GRA, Grasslands; CRO, Croplands; ENF, Evergreen Needleleaf Forest; DBF, Deciduous Broadleaf Forest; SHR, Shrubland.



1168

1169 **Fig. 7.** Changes in GPP for all and individual PFTs in the presence of different levels of
 1170 $[O_3]$ as simulated by the vegetation model. Simulations are performed at 40 NACP sites
 1171 with a fixed $[O_3]$ for either low or high O_3 sensitivity. The short blue lines show the
 1172 damages ranging from low to high O_3 sensitivity, with the blue points indicating the
 1173 average reductions. The simulation results are averaged for all the sites or for the sites
 1174 with the same PFT. The number of sites used for average is shown in the title bracket of
 1175 each subplot. The solid squares with lines show the results (mean plus uncertainty) based
 1176 on measurements reported by multiple literatures. These measurements are collected by
 1177 Lombardozzi et al. (2013) for all PFTs, Wittig et al. (2007) for evergreen needleleaf
 1178 forest (ENF) and deciduous broadleaf forest (DBF). For C4 grass or crop (CRO_C4), we
 1179 have collected data from Taylor et al. (2002) for spartina alterniflora and Grantz et al.
 1180 (2012) for sugarcane hybrids. For C3 grass or crop (GRA_C3), we have collected data
 1181 from Feng et al. (2008) for wheat, Foot et al. (1996) for colluna vulgaris, Mulchi et al.
 1182 (1992) for soybean, and Ishii et al. (2004) and Ainsworth (2008) for rice. The values for
 1183 rice are denoted in green and others in red. The author initials are indicated for the
 1184 corresponding studies.

1185

1186
1187
1188
1189
1190
1191
1192
1193
1194
1195
1196
1197
1198
1199
1200
1201
1202
1203
1204
1205
1206
1207
1208
1209
1210
1211
1212
1213
1214
1215
1216
1217
1218

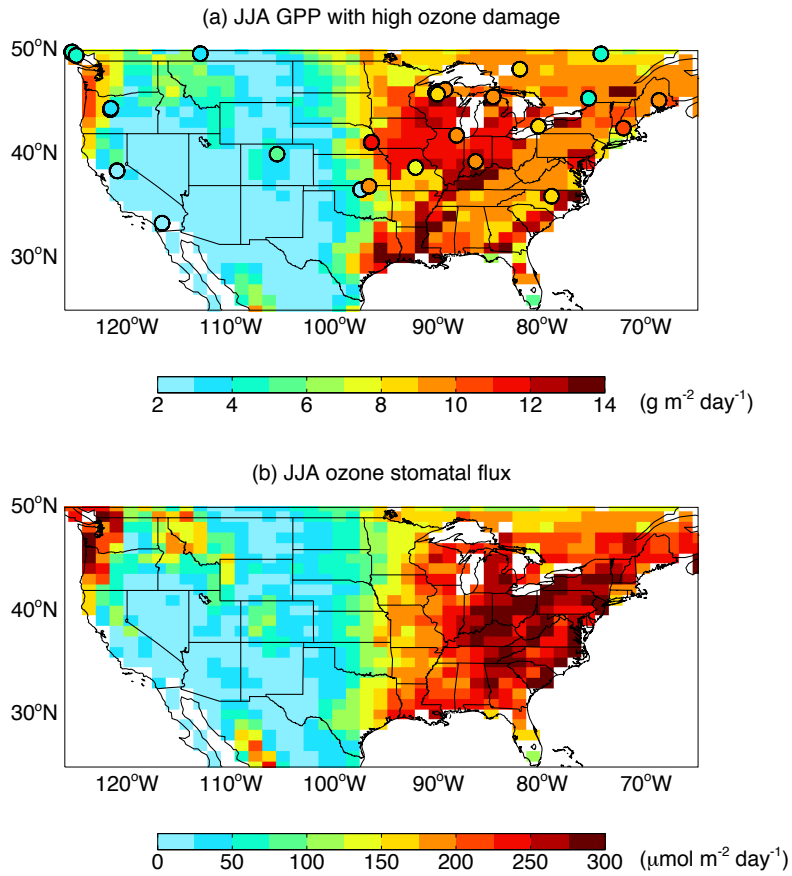


Fig. 8. Simulated summertime (a) O₃-exposed GPP and (b) O₃ stomatal flux over the U.S. The simulated GPP is overlaid with *in situ* measurements from NACP. The simulations are performed with land cover from ISLSCP and meteorological forcings from MERRA reanalyses. Please refer to Fig. S6 for separate results from simulations and observations.

1219
1220
1221
1222
1223
1224
1225
1226
1227
1228
1229
1230
1231
1232
1233
1234
1235
1236
1237
1238
1239
1240
1241
1242
1243
1244
1245
1246

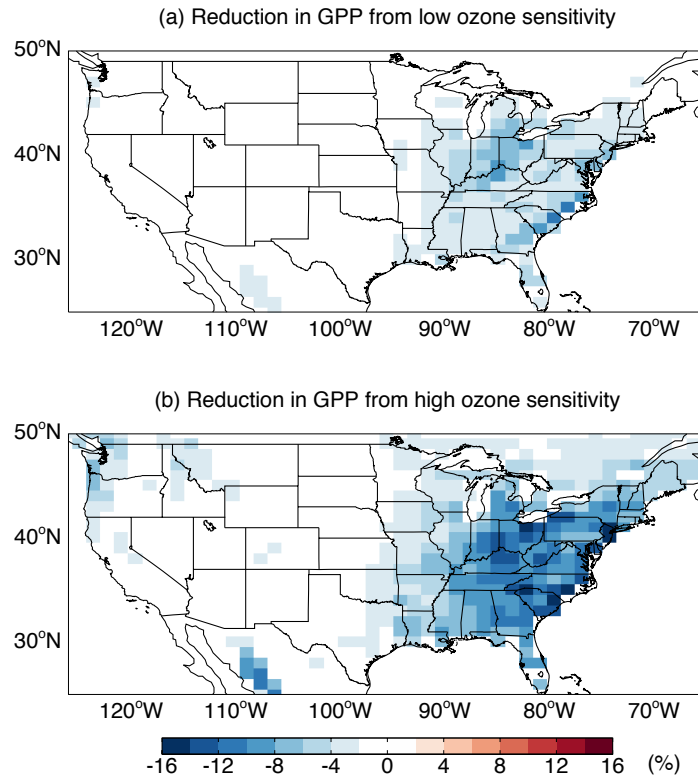
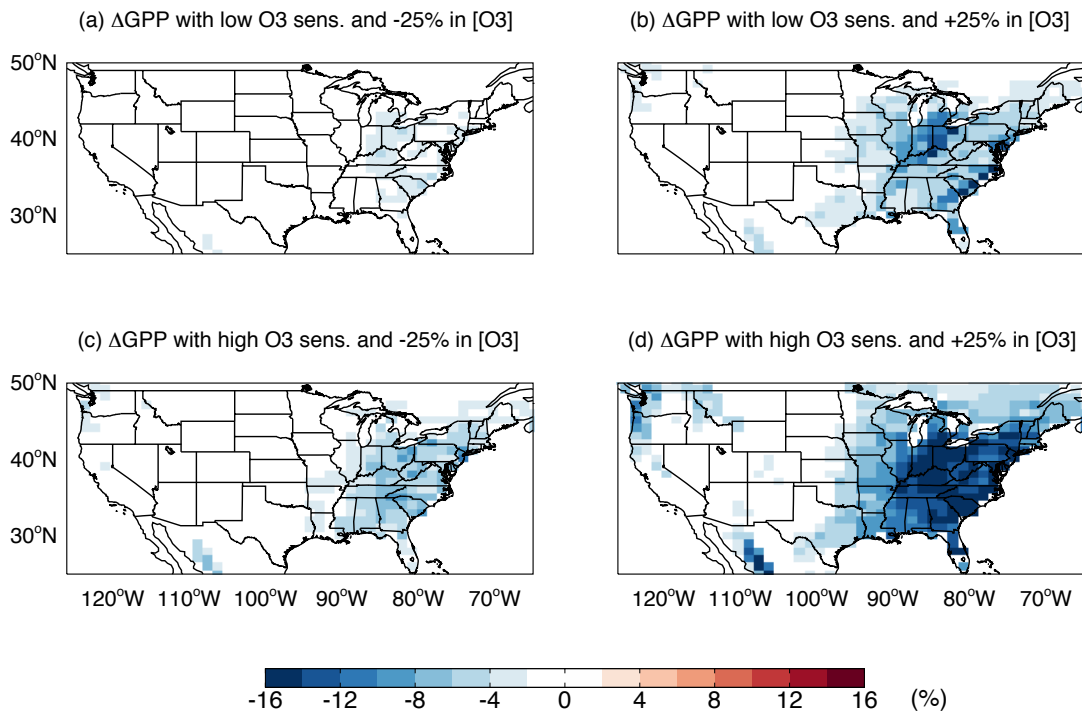


Fig. 9. Simulated reduction fraction in summer GPP in the U.S. due to (a) low and (b) high O_3 sensitivity for 1998-2007.

1247
1248
1249
1250
1251



1254
1255
1256
1257
1258
1259
1260
1261

Fig. 10. Simulated changes in summer GPP due to (a, c) 25% reduction or (b, d) 25% increase in [O₃] for (a, b) low or (c, d) high O₃ sensitivity.