

1 **Global assessment of climatic responses to the ozone-vegetation**
2 **interactions**

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12 **Abstract.** The coupling between surface ozone (O_3) and vegetation significantly
13 influences the regional to global climate. O_3 uptake by plant stomata inhibits the
14 photosynthetic rate and stomatal conductance, impacting evapotranspiration through
15 land surface ecosystems. Using a climate–vegetation–chemistry coupled model (the
16 NASA GISS ModelE2 coupled with the Yale Interactive terrestrial Biosphere, or
17 ModelE2-YIBs), we assess the global climatic responses to O_3 –vegetation interactions
18 during the boreal summer of the present day (2005–2014). High O_3 pollution reduces
19 stomatal conductance, resulting in warmer and drier conditions worldwide. The most
20 significant responses are found in the eastern U.S. and eastern China, where the surface
21 air temperature increases by $+0.33\pm 0.87$ °C and $+0.56\pm 0.38$ °C, respectively. These
22 temperature increases are accompanied by decreased latent heat and increased sensible
23 heat in both regions. The O_3 –vegetation interaction also affects atmospheric pollutants.
24 The surface maximum daily 8-hour average O_3 concentrations increase by $+1.46\pm 3.02$
25 ppbv in eastern China and $+1.15\pm 1.77$ ppbv in the eastern U.S. due to the O_3 -induced
26 inhibition of stomatal uptake. With reduced atmospheric stability following a warmer
27 climate, increased cloud cover but decreased relative humidity jointly reduce aerosol
28 optical depth by -0.06 ± 0.01 ($-14.67\pm 12.15\%$) over eastern China. This study suggests
29 that vegetation feedback should be considered for a more accurate assessment of
30 climatic perturbations caused by tropospheric O_3 .

31 **1 Introduction**

32 Tropospheric ozone (O₃), one of the most detrimental air pollutants (Myhre et al.,
33 2013), not only threatens human health (Norval et al., 2011; Nuvolone et al., 2018) but
34 also induces phytotoxic effects on vegetation (Mills et al., 2007; Pleijel et al., 2007).
35 When plants are exposed to certain levels of O₃, plant photosynthesis and stomatal
36 conductance are inhibited because of the O₃ oxidation of cells, enzymes, and
37 chlorophyll (Dizengremel, 2001; Fiscus et al., 2005; Jolivet et al., 2016). Consequently,
38 carbon assimilation in terrestrial ecosystems is limited (Yue and Unger, 2014; Oliver et
39 al., 2018), and the land–air exchange rates of water and heat fluxes are altered
40 (Lombardozzi et al., 2015).

41 Experimental studies have shown that excessive O₃ exposure reduces both plant
42 photosynthesis and stomatal conductance (Ainsworth et al., 2012; Lombardozzi et al.,
43 2013). The reduction rates are dependent on the O₃ stomatal fluxes as well as the
44 damage sensitivities, which vary among different vegetation types (Nussbaum and
45 Fuhrer, 2000; Karlsson et al., 2004; Pleijel et al., 2004). Several exposure-based indices,
46 such as accumulated hourly O₃ concentrations over a threshold of 40 ppb (AOT40) and
47 the sum of all hourly average concentrations (SUM00), are used to assess O₃-induced
48 vegetation damage (Fuhrer et al., 1997; Paoletti et al., 2007). In addition, the flux-
49 related POD_y method (phytotoxic O₃ dose above a threshold flux of y) is also widely
50 applied to consider the dynamic adjustment of stomatal conductance (Buker et al., 2015;
51 Sicard et al., 2016). Considering the variability of plant sensitivities, different O₃
52 damage schemes have been proposed to quantify the impacts of O₃ on land carbon
53 assimilation at regional to global scales (Anav et al., 2011; Lam et al., 2023; Lei et al.,
54 2020). For example, Sitch et al. (2007) calculated simultaneous damage to both
55 photosynthesis and stomatal conductance on the basis of instantaneous O₃ stomatal
56 uptake. In contrast, Lombardozzi et al. (2012) estimated decoupled reductions in plant
57 photosynthesis and stomatal conductance via different response relationships to
58 cumulative O₃ stomatal uptake. The application of different schemes has resulted in a
59 wide range of reductions in gross primary productivity (GPP) of 2–12% globally, with
60 regional hotspots of up to 20–30% (Lombardozzi et al., 2015; Unger et al., 2020; Zhou

61 et al., 2024).

62 O₃-induced inhibition of stomatal conductance decreases dry deposition and
63 consequently enhances surface O₃ concentrations (Clifton et al., 2020; Wesely and
64 Hicks, 2000; Zhang et al., 2006). Using the Sitch et al. (2007) scheme with high O₃
65 damage sensitivities in ModelE2-YIBs (NASA GISS ModelE2 coupled with the Yale
66 Interactive terrestrial Biosphere model), Gong et al. (2020) revealed that O₃-vegetation
67 interactions increased regional O₃ concentrations by 1.8 ppbv in the eastern U.S., 1.3
68 ppbv in Europe, and 2.1 ppbv in eastern China in 2010. In comparison, Sadiq et al.
69 (2017) reported consistently stronger feedback on O₃ concentrations in these polluted
70 regions via the scheme of Lombardozzi et al. (2012) embedded in the Community Earth
71 System Model (CESM). Moreover, the inclusion of online O₃-vegetation interactions
72 in numerical models will also result in a greater loss of simulated land carbon
73 assimilation due to the feedbacks of both ecosystems and surface O₃. This is attributable
74 to several factors. On the one hand, O₃ damage to leaf photosynthesis inhibits plant
75 growth and decreases the leaf area index (LAI), leading to a greater reduction in GPP
76 than in simulations without LAI changes (Yue et al., 2020). On the other hand, O₃
77 enhancement due to vegetation feedback may cause additional vegetation damage and
78 result in further GPP losses (Lei et al., 2021). As a result, O₃-vegetation interactions
79 should be considered in global estimates of O₃ damage to ecosystem functions.

80 In addition to affecting surface O₃, O₃-vegetation interactions can also alter water
81 and energy exchange between the land and atmosphere through the modulation of
82 stomatal conductance. For example, Lombardozzi et al. (2015) used the Community
83 Land Model (CLM) and estimated that the cumulative uptake of O₃ by leaves resulted
84 in a reduction of 2.2% in transpiration but an increase of 5.4% in runoff globally. Arnold
85 et al. (2018) used CESM and reported that plant exposure to O₃ could decrease land-
86 air moisture fluxes and atmospheric humidity, which further reduced shortwave cloud
87 forcing in polluted regions and induced widespread surface warming up to +1.5 K. Two
88 recent studies utilized the WRF-Chem model and revealed considerable warming and
89 associated meteorological perturbations due to O₃-vegetation interactions in China
90 (Zhu et al., 2022; Jin et al., 2023). However, all these modeling studies applied the same

91 O₃ vegetation damage scheme proposed by Lombardozzi et al. (2012). It is necessary
92 to assess the climatic responses to O₃–vegetation interactions via different schemes to
93 explore robust responses and associated uncertainties.

94 In this study, we quantified the global impacts of O₃–vegetation interactions on
95 climatic conditions and surface air pollutants during the 2010s via ModelE2-YIBs (Yue
96 and Unger, 2015). This fully coupled framework was implemented with the
97 semimechanistic O₃ damage scheme proposed by Sitch et al. (2007), which calculates
98 aggregated O₃ damage to photosynthesis on the basis of varied sensitivities to
99 instantaneous stomatal O₃ uptake across eight plant functional types (PFTs). We
100 performed sensitivity experiments to quantify the responses of surface air temperature
101 and precipitation to O₃–vegetation interactions. The feedbacks to aerosols and O₃
102 concentrations were also examined.

103

104 **2 Method**

105 **2.1 Model descriptions**

106 ModelE2-YIBs is a fully coupled climate–carbon–chemistry model that combines
107 the NASA GISS ModelE2 with the YIBs vegetation model. ModelE2 is a general
108 circulation model with a horizontal resolution of 2°×2.5° in latitude and longitude and
109 40 vertical layers up to 0.1 hPa. It dynamically simulates gas-phase chemistry (NO_x,
110 HO_x, O_x, CO, CH₄, and NMVOCs), aerosols (sulfate, nitrate, black and organic carbon,
111 dust, and sea salt), and their interactions (Menon and Rotstayn, 2006). Both the physical
112 and chemical processes are calculated every 0.5 h, and the radiation module is called
113 every 2.5 h. The radiation module includes direct and indirect aerosol radiative effects
114 and accounts for the absorption of multiple greenhouse gases (GHGs). For cloud optical
115 parameters, Mie scattering, ray tracing, and matrix theory are used (Schmidt et al.,
116 2006). The model outperforms 20 other IPCC-class climate models in simulating
117 surface solar radiation (Wild et al., 2013) and has been extensively validated for
118 meteorological and hydrological variables against observations and reanalysis data
119 (Schmidt et al., 2014).

120 The YIBs model employs the well-established Farquhar model for leaf

121 photosynthesis and the Ball-Berry model for stomatal conductance (Farquhar et al.,
 122 1980; Ball et al., 1987) as follows:

$$A_{tot} = \min(J_c, J_e, J_s) \quad (1)$$

123 Here, the total leaf photosynthesis, denoted as A_{tot} ($\mu\text{mol m}^{-2} [\text{leaf}] \text{s}^{-1}$), is calculated
 124 considering both C_3 (Collatz et al., 1991) and C_4 plants (Collatz et al., 1992). A_{tot} is
 125 derived from the minimum value of the constraints. The rate of carboxylation limited
 126 by ribulose-1,5-bisphosphate carboxylase (Rubisco) is J_c :

$$J_c = \begin{cases} V_{cmax} \left(\frac{c_i - \Gamma_*}{c_i + K_c(1 + O_i/K_o)} \right) & \text{for } C_3 \text{ plant} \\ V_{cmax} & \text{for } C_4 \text{ plant} \end{cases} \quad (2)$$

127 The carboxylation rate restricted by the availability of light is J_e :

$$J_e = \begin{cases} a_{leaf} \times PAR \times \alpha \times \left(\frac{c_i - \Gamma_*}{c_i + 2\Gamma_*} \right) & \text{for } C_3 \text{ plant} \\ a_{leaf} \times PAR \times \alpha & \text{for } C_4 \text{ plant} \end{cases} \quad (3)$$

128 The export-limited rates for C_3 plants and the phosphoenolpyruvate carboxylase
 129 (PEPC)-limited rates of carboxylation for C_4 plants are represented by J_s :

$$J_s = \begin{cases} 0.5 V_{cmax} & \text{for } C_3 \text{ plant} \\ K_s \times V_{cmax} \times \frac{c_i}{P_{atm}} & \text{for } C_4 \text{ plant} \end{cases} \quad (4)$$

130 In these functions, V_{cmax} ($\mu\text{mol m}^{-2} \text{s}^{-1}$) is the maximum carboxylation capacity. c_i
 131 and O_i (Pa) represent the internal leaf CO_2 and oxygen partial pressure, respectively.
 132 Γ_* (Pa) denotes the CO_2 compensation point, whereas K_c and K_o (Pa) are Michaelis–
 133 Menten constants for the carboxylation and oxygenation of Rubisco, respectively. The
 134 parameters Γ_* , K_c , and K_o vary with temperature on the basis of the sensitivity of the
 135 vegetation to temperature (Q_{10} coefficient). PAR ($\mu\text{mol m}^{-2} \text{s}^{-1}$) is the absorbed
 136 photosynthetically active radiation, a_{leaf} is the leaf-specific light absorbance that
 137 considers sunlit and shaded leaves, and α is the quantum efficiency. P_{atm} (Pa)
 138 represents the ambient pressure. K_s is set to 4000 as a constant following Oleson et al.
 139 (2010) to limit photosynthesis to C_4 plants that become saturated at lower CO_2
 140 concentrations.

141 The stomatal conductance (g_s , $\text{mol} [\text{H}_2\text{O}] \text{m}^{-2} \text{s}^{-1}$) is linked to the variations in
 142 A_{tot} with several parameters, such as the dark respiration rate (R_d , $\mu\text{mol m}^{-2} \text{s}^{-1}$),
 143 relative humidity (RH), and CO_2 concentration at the leaf surface (c_s):

$$g_s = m \frac{(A_{tot} - R_d) \times RH}{c_s} + b \quad (5)$$

144 The model simulates the biophysical processes of eight PFTs, including tundra, C₃/C₄
 145 grass, shrubland, deciduous broadleaf forest, evergreen broadleaf forest, evergreen
 146 needleleaf forest, and cropland. Different values are assigned to parameters *m* and *b* for
 147 each PFT (Table S1). The carbon taken up by the leaf then accumulates and is allocated
 148 to different organs to support plant development, resulting in dynamic changes in the
 149 LAI and tree growth.

150

151 2.2 O₃-vegetation damage scheme

152 The YIBs model employs a semimechanistic parameterization proposed by Sitch
 153 et al. (2007) to estimate the impact of O₃ on photosynthesis through stomatal uptake.
 154 The scheme applies an undamaged factor (*F*) (nmol m⁻² s⁻¹) to both *A_{tot}* and *g_s* as
 155 follows:

$$A_{totd} = A_{tot} \cdot F \quad (6)$$

$$g_{sd} = g_s \cdot F \quad (7)$$

156 where *A_{totd}* and *g_{sd}* are the unaffected photosynthesis and stomatal conductance,
 157 respectively. The factor *F* is defined as follows:

$$F = 1 - a_h \cdot \max [F_{O_3} - F_{O_3,crit}, 0.0] \quad (8)$$

158 where *a_h* (mmol m⁻² s⁻¹) is the high O₃ sensitivity coefficient, calibrated by Sitch et
 159 al. (2007) on data from field observations by Karlsson et al. (2004) and Pleijel et al.
 160 (2004) to represent the ‘high’ sensitivity of the relative species of each PFT. *F_{O₃,crit}*
 161 (nmol m⁻² s⁻¹) is the specific threshold for O₃ damage, both of which vary with
 162 vegetation type (Table S1):

$$F_{O_3} = \frac{[O_3]}{R_a + \left[\frac{k_{O_3}}{g_{sd}} \right]}, \quad (9)$$

163 where *[O₃]* represents surface O₃ concentrations, and *R_a* (s m⁻¹) represents
 164 aerodynamic resistance, which expresses the turbulent transport efficiency in
 165 transferring sensible heat and water vapor between the land surface and a reference
 166 height. The constant *k_{O₃}*=1.67 is the ratio of stomatal resistance to O₃, which is
 167 estimated on the basis of the theoretical stomatal resistance to water (Laisk et al., 1989).

168 When plants are exposed to $[O_3]$ (Eq. 9), A_{tot} and g_s decrease (Eq. 6 and Eq. 7) if
169 the excess O_3 enters the leaves (Eq. 8). The increased stomatal resistance protects plants
170 by reducing the O_3 uptake of stomata. Consequently, the damage scheme describes both
171 changes in the photosynthetic rate and stomatal conductance.

172

173 **2.3 Experiments**

174 To explore the coupled O_3 -vegetation effect, we performed two simulations using
175 the ModelE2-YIBs model. The control experiment “O3_offline” was conducted
176 without O_3 damage to vegetation. For comparison, the sensitivity experiment
177 “O3_online” included online O_3 -vegetation interactions with high O_3 sensitivity. For
178 both experiments, the anthropogenic emissions from 2010 (the average of 2005–2014)
179 for 8 species (BC, OC, CO, NH_3 , NO_x , SO_2 , alkenes, and paraffin) from 8 economic
180 sources (agriculture, energy, industry, transportation, residential, solvent, waste, and
181 international shipping) and biomass burning sources were collected from the Coupled
182 Model Intercomparison Project phase 6 (CMIP6) (van Marle et al., 2017; Hoesly et al.,
183 2018). The ensemble means of the monthly sea surface temperature (SST) and sea ice
184 concentration (SIC) simulated by 21 CMIP6 models during the time period of 2005–
185 2014 were employed as the boundary conditions. The cover fractions of 8 PFTs (Fig.
186 S1) fixed at 2010 were adopted from the land use harmonization (LUH2) dataset (Hurtt
187 et al., 2020). For each time-slice simulation, the model was run for 30 years with all the
188 input data fixed, and the first 10 years were used as the spin-up period. We calculated
189 the average of the last 20 years and focused on the boreal summer season (June–July–
190 August, JJA), when the interaction of vegetation and surface O_3 reaches its annual
191 maximum (Fig. S3). To show the uncertainty introduced by the internal variability of
192 the model, all the related global/regional values are denoted as mean/sum \pm standard
193 deviation of the last 20 model years. We explored the climatic responses to O_3 -
194 vegetation interactions as the differences between “O3_online” and “O3_offline” at the
195 global scale, with a focus on hotspot regions, such as the eastern U.S. (30–40°N, 80–
196 90°W) and eastern China (22.5–38°N, 106–122°E).

197

198 2.4 Data for model evaluation

199 We evaluated the simulated air pollutants, carbon fluxes, and meteorological
200 variables from the ‘O3_offline’ simulation against observational and reanalysis datasets.
201 The worldwide observations of the maximum daily 8-hour average O₃ (MDA8 O₃)
202 concentrations were collected from three regional networks: the Air Quality Monitoring
203 Network operated by the Ministry of Ecology and Environment (AQMN-MEE) in
204 China, the Clean Air Status and Trends Network (CASTNET) in the U.S., and the
205 European Monitoring and Evaluation Programme (EMEP) in Europe. The observations
206 used for validation beyond China, sourced from Sofen et al. (2016), are averaged over
207 the period 2005–2014. This dataset encompasses 7288 station records worldwide and
208 excludes the uncertainty associated with high mountain-top sites. For AQMN-MEE, the
209 mean value of 2014–2018 was used because it was established in 2013. The simulated
210 aerosol optical depth (AOD) and LAI were validated using satellite-based data from the
211 Moderate Resolution Imaging Spectroradiometer (MODIS) retrievals collection 5
212 (Remer et al., 2005) (<http://modis.gsfc.nasa.gov/>) averaged over the years 2005–2014.
213 The simulated GPP was evaluated against the data product upscaled from the
214 FLUXNET eddy covariance measurements for 2009–2011 (Jung et al., 2011). The daily
215 temperature at 2 m (T_{2m}) from 2005–2014 was obtained from the National Centers for
216 Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR)
217 reanalysis 1 (NCEP1) (Kalnay et al., 1996). For precipitation, we used the monthly data
218 averaged from 2005–2014 from the Global Precipitation Climatology Project (GPCP)
219 (Huffman et al., 1997; Adler et al., 2018). All these datasets were interpolated to the
220 same resolution as the ModelE2-YIBs model. The root-mean-square error (RMSE) and
221 normalized mean bias (NMB) were applied to quantify the deviations of the simulations
222 from the observations:

$$223 \quad RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (S_i - O_i)^2} \quad (10)$$

$$NMB = \frac{\sum_{i=1}^n (S_i - O_i)}{\sum_{i=1}^n O_i} \times 100\% \quad (11)$$

224 Here, S_i and O_i represent the simulated and observed values, respectively; *n* denotes the
225 total number of grid points used in the comparisons.

226

227 **3. Results**

228 **3.1 Control simulation and model evaluations**

229 We first evaluated the air pollutants simulated by the control simulation
230 (O3_offline) of the ModelE2-YIBs model (Fig. 1). Over a total of 503 grids with site-
231 level O₃ measurements (Fig. 1b), the model replicated both the magnitude and spatial
232 distribution of MDA8 O₃, with a correlation coefficient (r) of 0.59 and NMB of -2.54%
233 (Fig. 1c). The simulated summertime surface MDA8 O₃ concentration was high in
234 regions with high anthropogenic emissions, such as western Europe and eastern China
235 (Ohara et al., 2007), as well as in central Africa, which has frequent fire emissions (van
236 der Werf et al., 2017). At the global scale, the model yielded an average MDA8 O₃
237 concentration of 43.93 ppbv, and observations revealed an average of 44.72 ppbv over
238 the same grids. However, the model overestimated the concentrations over the North
239 China Plain and slightly underestimated them over the U.S., likely due to biases in the
240 emission inventories and the predicted climate that drive O₃ production. The simulated
241 AOD at 550 nm by O3_offline (Fig. 1d) showed a spatial pattern similar to that of the
242 satellite retrievals (Fig. 1e), with $r = 0.75$ and an NMB of -7.35% globally (Fig. 1f).
243 Both simulations and observations revealed AOD hotspots over North Africa and the
244 Middle East, where dust emissions dominate, and in northern India and eastern China,
245 where anthropogenic emissions are high (Feng et al., 2020).

246 We then evaluated the simulated GPP and LAI via a control experiment for the
247 boreal summer period (Fig. 2). The observations revealed GPP hotspots over boreal
248 forests, such as those in the eastern U.S., Eurasia, and East Asia, and tropical forests,
249 such as those in the Amazon, Central Africa, and Indonesia (Fig. 2b). The seasonal total
250 GPP was estimated to be 41.63 Pg[C], which accounted for 35% of the annual amount.
251 The simulations captured the observed GPP pattern at the global scale, with $r = 0.64$
252 and NMB = -7.81% across 2581 grids (Fig. 2c), with underestimations in the tundra
253 area and slight overestimations in the tropical rainforest and evergreen forest regions.
254 The model simulated a seasonal total GPP of 38.69 Pg[C], equivalent to 34% of the
255 annual amount. The simulated LAI showed patterns similar to those of GPP (Fig. 2d)

256 and resembled the observed LAI (Fig. 2e), with a spatial correlation of $r = 0.79$ and a
257 low NMB = -5.43% across 4435 grids globally (Fig. 2f).

258 We further validated the simulated meteorology from O3_offline (Fig. S2). For the
259 surface air temperature, the model (Fig. S2a) reproduced the observed (Fig. S2b) pattern,
260 with an RMSE of 3.21 °C and an r of 0.99 compared with the observations (Fig. S2c).
261 For precipitation, the simulation (Fig. S2d) captured the observed spatial pattern (Fig.
262 S2e), with NMB = 17.26% and $r = 0.75$ (Fig. S2f). Overall, the model captured the
263 spatial characteristics and magnitudes of air pollutants, biospheric parameters, and
264 meteorological fields, making it a valuable tool for studying O₃-vegetation interactions.
265

266 **3.2 O₃ damage to terrestrial ecosystems**

267 We assessed the damaging effects of surface O₃ on ecosystems due to online O₃-
268 vegetation interactions (Fig. 3). The impacts of O₃ on biospheric variables were mainly
269 located in regions characterized by abundant vegetation cover and elevated O₃
270 concentrations. On the global scale, O₃ induced a GPP reduction of -1.80 ± 0.61 PgC yr⁻¹
271 ¹ ($-4.69 \pm 1.56\%$, Fig. 3a). This deleterious effect was more pronounced in specific
272 regions, notably eastern China and eastern U.S., with significant GPP declines of -
273 $25.40 \pm 1.90\%$ and $-20.14 \pm 5.02\%$, respectively, under high O₃ sensitivity conditions (Fig.
274 3a and Table S2). Moreover, stomatal conductance significantly decreased in the middle
275 latitudes of the Northern Hemisphere (Fig. 3b). The most substantial relative change of
276 $-30.62 \pm 4.30\%$ was observed in eastern China, followed by $-25.65 \pm 9.32\%$ in the eastern
277 U.S. (Fig. 3b and Table S2). Although there are positive responses in some regions, they
278 are not dominant and are hardly significant. These values were stronger than those for
279 GPP (Fig. 3a), likely because of the climatic feedback to O₃-vegetation interactions.
280 The opening of the plant stoma plays a crucial role in regulating energy and water
281 exchange between the land surface and the atmosphere. The inhibition of stomatal
282 conductance by surface O₃ leads to a warmer (Fig. 4a) and drier (Fig. 4b) climate in
283 those hotspot regions, resulting in even stronger inhibitory effects on stomatal
284 conductance. Following the changes in GPP, the global LAI on average decreased by
285 0.01 ± 0.01 m² m⁻² ($-0.62 \pm 0.84\%$), with regional maxima of $-4.53 \pm 1.14\%$ in eastern

286 China and $-5.87\pm 3.11\%$ in the eastern U.S. (Table S2).

287

288 **3.3 Global climatic responses to O₃–vegetation interactions**

289 In response to the O₃-induced inhibition of stomatal conductance, surface air
290 temperature increased by 0.05 ± 0.20 °C (Fig. 4a), whereas precipitation decreased by -
291 0.01 ± 0.03 mm day⁻¹ (Fig. 4b) at the global scale. The most significant change was the
292 warming of 0.56 ± 0.38 °C and precipitation reduction of -0.79 ± 1.05 mm day⁻¹ (-
293 $16.18\pm 20.38\%$) in eastern China (Table S3), followed by the greatest inhibition of
294 stomatal conductance (Fig. 3b). Such warming and rainfall deficits also appeared in the
295 eastern U.S. and western Europe, where the O₃–vegetation interactions were notable.
296 The O₃-induced inhibition of stomatal conductance decreased the latent heat flux (Fig.
297 4e) and consequent precipitation (Fig. 4b) in those hotspot regions. Moreover, the
298 reduced latent heat flux promoted higher surface air temperatures (Fig. 4a), resulting in
299 an increase in the sensible heat flux (Fig. 4f). Such warming has also been reported in
300 field experiments, where relatively high O₃ exposure resulted in noticeable increases in
301 canopy temperature along with reductions in transpiration (Bernacchi et al., 2011;
302 VanLoocke et al., 2012). Globally, temperature and precipitation showed patchy
303 responses with both positive and negative anomalies, suggesting that the regional
304 hotspots of O₃-induced meteorological changes propagate to surrounding areas through
305 atmospheric perturbations.

306 We further examined the changes in air humidity and cloud cover. The surface
307 relative humidity decreased by $-0.18\pm 0.53\%$ globally, with a similar pattern as that of
308 precipitation (Fig. 4c). The most significant reductions occurred over eastern China and
309 the eastern U.S., where both warming (Fig. 4a) and rainfall deficit (Fig. 4b) contributed
310 to drought. However, in adjacent regions, such as northern China and the central U.S.,
311 both rainfall and surface relative humidity increased. These changes were associated
312 with the regional increase in cloud cover (Fig. 4d). The sensible heat flux increased by
313 6.3 ± 5.4 W m⁻² ($16.54\pm 15.59\%$) and 7.12 ± 3.86 W m⁻² ($25.46\pm 14.71\%$) in the eastern
314 U.S. and eastern China, respectively, suggesting the transfer of thermal energy from the
315 land to the atmosphere via O₃–vegetation interactions (Fig. 4f and Table S3). The

316 warming effect further triggered anomalous updrafts in the lower troposphere,
317 represented by changes in vertical velocity (Fig. 5), leading to enhanced convection,
318 reduced atmospheric stability, and consequently an increase in low-level cloud cover
319 (Fig. 4d). However, despite the usual cooling effect associated with increased cloud
320 cover due to reductions in radiation, in regions predominantly influenced by O₃-
321 vegetation interactions, this cooling effect was outweighed by O₃-induced warming
322 through the inhibition of stomatal conductance. Therefore, temperatures exhibited an
323 overall increase of 0.56 ± 0.38 °C in eastern China and 0.33 ± 0.87 °C in the eastern U.S.
324 (Table S3).

325

326 **3.4 Changes in air pollution caused by O₃-vegetation interactions**

327 Changes in surface water and heat fluxes induced by O₃-vegetation interactions
328 could feed back to affect air pollutants, such as O₃ and aerosols. As shown in Fig. 6a
329 and Table S4, surface MDA8 O₃ concentrations increased by 1.46 ± 3.02 ppbv in eastern
330 China and 1.15 ± 1.77 ppbv in the eastern U.S. due to decreased dry deposition following
331 O₃ inhibition of stomatal conductance. This indicates that high contemporary O₃
332 pollution may worsen air quality through O₃-vegetation interactions. However,
333 negative O₃ changes were predicted in the central U.S. and western China, where
334 increased rainfall dampened O₃ through chemical reactions and wet deposition. On a
335 global scale, surface MDA8 O₃ concentrations showed a limited increase of 0.03 ± 0.4
336 ppbv because of the offset between positive and negative feedbacks. The increase in O₃
337 concentrations in polluted regions may exacerbate the warming effect of O₃ as a
338 greenhouse gas and cause additional damage to vegetation. For example, the effects of
339 offline O₃ damage on GPP in eastern China and the eastern US were simulated to be -
340 0.52 ± 0.03 Pg[C] ($-24.98\pm 0.91\%$) and -0.17 ± 0.02 Pg[C] ($-16.71\pm 1.16\%$), respectively,
341 which are smaller than those induced by O₃-vegetation interactions (Table S2).

342 Aerosols also exhibited evident changes in response to O₃-vegetation interactions.
343 The AOD significantly decreased over hotspot regions, such as eastern China and the
344 eastern U.S. (Fig. 6b). In the ModelE2-YIBs model, sulfate was especially sensitive to
345 clouds, which could enhance aerosol scavenging through cloud water precipitation

346 (Koch et al., 2006). The large increase in cloud cover removed sulfate more efficiently
347 than the other aerosol species did, leading to an average decrease of $-1.94 \pm 1.67 \mu\text{g m}^{-3}$
348 ($-8.52 \pm 6.88\%$) in the $\text{PM}_{2.5}$ loading over eastern China (Fig. S4 and Table S4).
349 Moreover, the reduction in surface relative humidity (Fig. 4c) in the regions with strong
350 O_3 -vegetation interactions limited the hygroscopic growth of aerosols, leading to a
351 more noticeable decrease in AOD (Petters and Kreidenweis, 2007; Pitchford et al., 2007)
352 by -0.06 ± 0.05 ($-14.67 \pm 16.75\%$) in eastern China (Table S4). Similar aerosol changes
353 were found in the eastern U.S. but with smaller reductions in $\text{PM}_{2.5}$ by $-0.27 \pm 0.36 \mu\text{g}$
354 m^{-3} ($-6.01 \pm 7.9\%$) and AOD by -0.01 ± 0.01 ($-8.15 \pm 9.38\%$) (Table S4). In addition to the
355 key O_3 -vegetation coupling regions, positive but insignificant changes in AOD were
356 predicted, leading to moderate AOD changes at the global scale (Fig. 6b).

357

358 **4. Discussion and conclusions**

359 We examined O_3 -vegetation feedback to climate and air pollution in the 2010s via
360 the fully coupled climate-carbon-chemistry model ModelE2-YIBs. During boreal
361 summer, surface O_3 resulted in strong damage to GPP and inhibited stomatal
362 conductance with regional hotspots over eastern China and the eastern U.S.
363 Consequently, surface transpiration was weakened, leading to decreased latent heat
364 fluxes and relative humidity but increased surface air temperature. Moreover, surface
365 warming increased cloud cover by reducing atmospheric stability. However, the
366 increase in cloud cover decreased the surface temperature and promoted precipitation
367 outside the key regions with intense O_3 -vegetation interactions. The O_3 -induced
368 inhibition of stomatal conductance resulted in a localized increase in O_3 concentration.
369 In contrast, the increased cloud cover and decreased relative humidity jointly reduced
370 the AOD in hotspot regions. At the global scale, the mean changes in both climate and
371 air pollution were moderate because of the offset between the changes with opposite
372 signs.

373 Our predictions of the changes in water/heat fluxes caused by O_3 -vegetation
374 interactions were consistent with those of previous studies (Lombardozzi et al., 2015;
375 Arnold et al., 2018; Gong et al., 2020). For example, simulations by Lombardozzi et al.

376 (2015) revealed that surface O₃ reduces global GPP by 8-12% and transpiration by 2-
377 2.4%, with regional reductions of up to 20% for GPP and 15% for transpiration in
378 eastern China and the U.S. These changes are generally consistent with our results,
379 although we predicted greater reductions in transpiration than in GPP due to O₃-
380 vegetation interactions. Using the same scheme as Lombardozzi et al. (2015), Sadiq et
381 al. (2017) reported that O₃-vegetation coupling induced surface warming of 0.5–1 °C
382 and O₃ enhancement of 4–6 ppbv in eastern China and eastern U.S. The magnitude of
383 these responses was much stronger than our predictions, likely because they considered
384 the accumulation effect of O₃. In contrast, regional simulations by Jin et al. (2023)
385 revealed that O₃-vegetation coupling led to increases in temperature of up to 0.16 °C
386 and surface O₃ of up to 0.6 ppbv in eastern China, both of which were smaller than our
387 predictions. The damage scheme they use, which depends on cumulative O₃ uptake,
388 omits the difference in impact on sunlit or shaded leaves and overestimates the O₃
389 damage to GPP compared with the scheme we use, which considers transient O₃ flux
390 (Cao et al., 2024). The discrepancies in O₃-vegetation feedbacks when the same O₃
391 damage schemes were used revealed uncertainties in the climate and chemistry models.
392 Our predictions were within the range of previous estimates for both climatic and O₃
393 changes.

394 There were several limitations in our simulated O₃-vegetation interactions. First,
395 the semimechanistic O₃ damage scheme we used in the present study linked damage to
396 photosynthesis with damage to stomatal conductance (Sitch et al., 2007), leading to a
397 greater percentage of inhibition of stomatal conductance than of photosynthesis,
398 considering O₃-vegetation feedbacks. However, some observations have shown that
399 damage to stomatal conductance occurs more slowly and might not be proportional to
400 the decline in photosynthetic rates (Gregg et al., 2006; Lombardozzi et al., 2012).
401 Second, observations have shown large variability in plant sensitivity to O₃ damage.
402 The Sitch et al. (2007) scheme employs low to high ranges of sensitivity to indicate
403 interspecific variabilities. In this study, we employed only high O₃ sensitivity to explore
404 the maximum responses. The possible uncertainties due to varied O₃ damage
405 sensitivities deserve further investigation. Third, large-scale observations were not

406 available to validate the simulated regional to global responses of climate and air
407 pollutants. The O₃ vegetation damage scheme has been extensively validated against
408 site-level measurements of both photosynthesis (Yue and Unger, 2018) and stomatal
409 conductance (Yue et al., 2016). However, we were conservative about the derived
410 global responses given that previous studies have shown large discrepancies when the
411 same O₃ damage scheme is used but have been implemented in different climate and/or
412 chemistry models (Lombardozzi et al., 2015; Sadiq et al., 2017; Jin et al., 2023).
413 Furthermore, the 2°×2.5° resolution of the current version of the ModelE2-YIBs model
414 has limitations due to the high computational demands. However, high-resolution
415 models exhibit improved simulations of extreme events (Chang et al., 2020; Ban et al.,
416 2021), which have certain effects on O₃–vegetation interactions (Mills et al., 2016; Lin
417 et al., 2020). While chemical transport models with relatively coarse resolutions can
418 increase biases in simulated air pollutants, they still capture large-scale patterns similar
419 to those of fine-resolution results and compare reasonably well to observational data
420 (Wang et al., 2013; Li et al., 2016; Lei et al., 2020). Moreover, we omit the slow climatic
421 feedback caused by air–sea interactions in the simulations. Studies have revealed that
422 these interactions may result in different climatic perturbations from those in
423 simulations with fast responses of the land surface alone (Yue et al., 2011). A dynamic
424 ocean model would enrich future research. Moreover, this study did not isolate the
425 different impacts of aerosols, even though the radiation module included both direct
426 and indirect radiative effects. We will investigate this further in the future by identifying
427 the main processes involved.

428 Despite these uncertainties, our simulations revealed considerable changes in both
429 climate and air pollutants in response to O₃–vegetation interactions. The most intense
430 warming, dryness, and O₃ enhancement were predicted in eastern China and the eastern
431 U.S., affecting the regional climate and threatening public health for these top two
432 economic centers. In contrast, for the first time, we revealed a reduction in aerosol
433 loading in hotspot regions, suggesting both positive and negative effects on air
434 pollutants via O₃–vegetation feedback. Such interactions should be considered in Earth
435 system models to better project future changes in climate and air pollutants following

436 anthropogenic interventions to both O₃ precursor emissions and ecosystem functions.
437

438 **Data Availability**

439 The observational data and model outputs that support the findings in this study are
440 available from the corresponding authors upon reasonable request.

441

442 **Author contributions**

443 XY conceived the project. XZ performed the model simulations, conducted the analysis
444 and wrote the draft manuscript. XY, CT and XL assisted in the interpretation of the
445 results and contributed to the discussion and improvement of the paper.

446

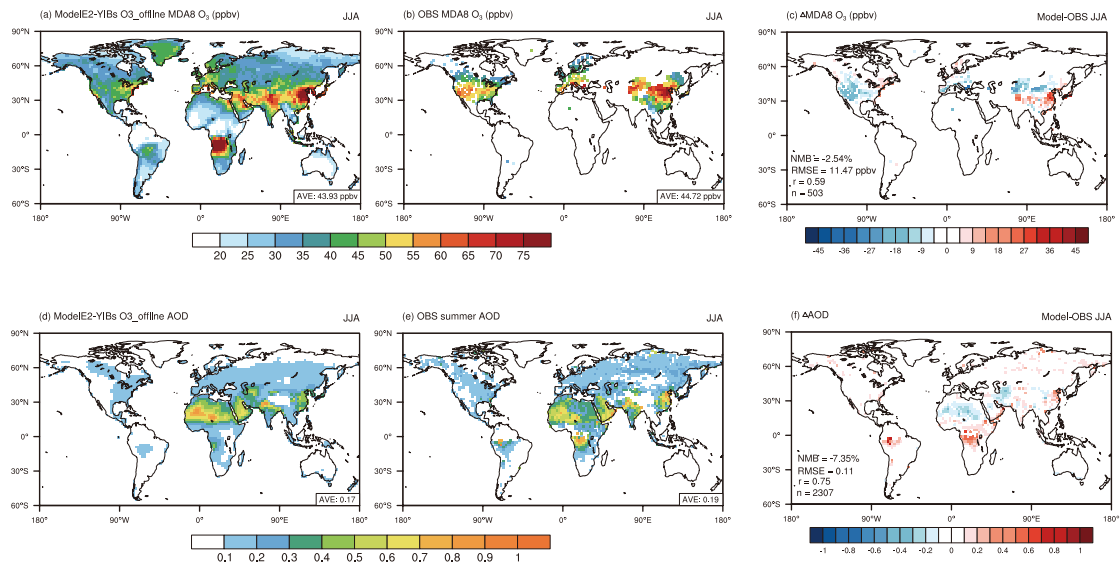
447 **Competing interests**

448 The authors declare that they have no conflicts of interest.

449

450 **Acknowledgments**

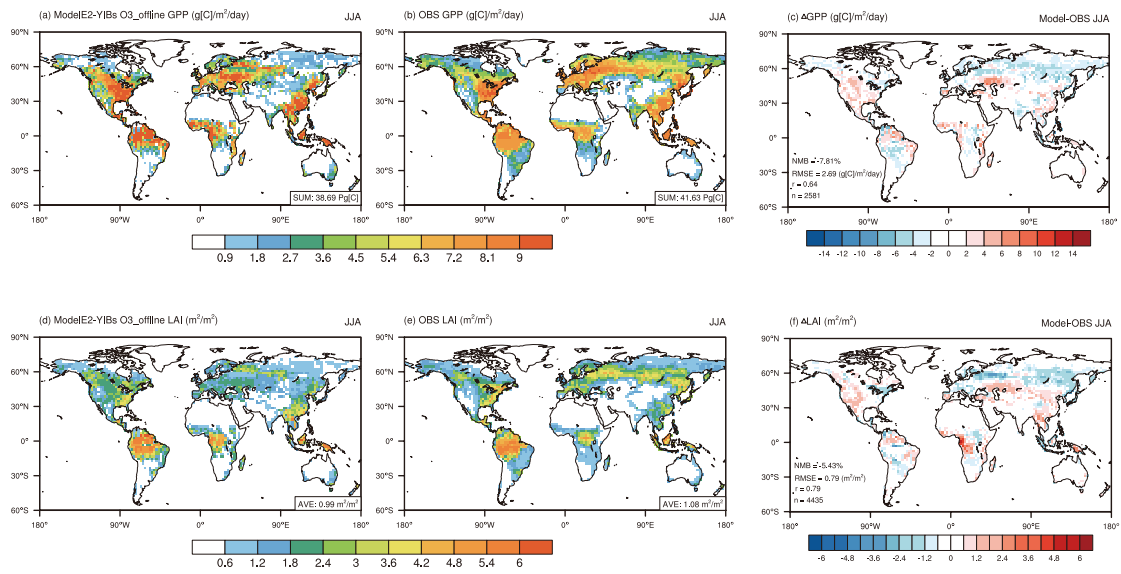
451 This study was jointly funded by the National Key Research and Development Program
452 of China (no. 2023YFF0805404) and the National Natural Science Foundation of China
453 (no. 42293323).



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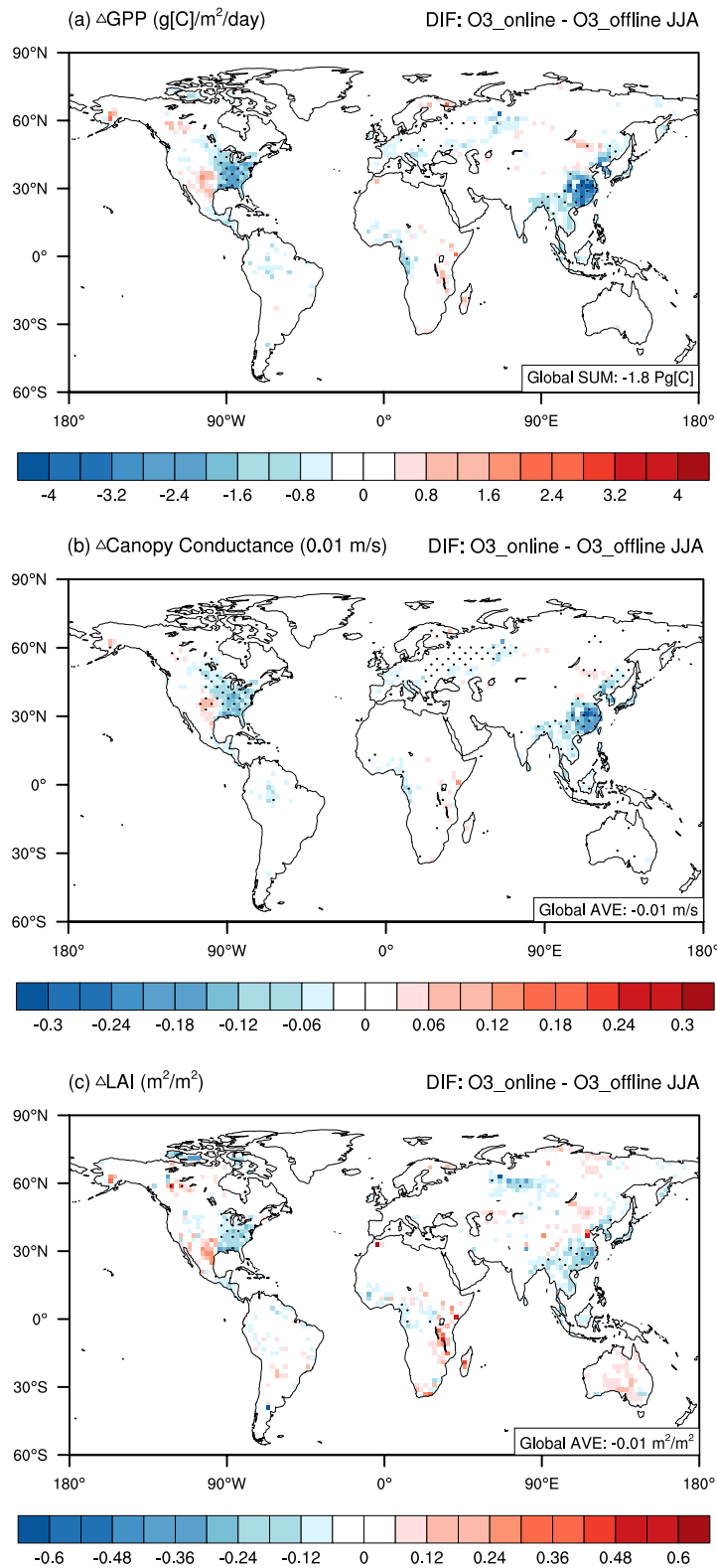
455 **Figure 1.** Evaluation of the present-day boreal summertime (June–August) air
 456 pollutants simulated by the ModelE2-YIBs model. The MDA8 O₃ (a-c) and AOD (d-f)
 457 from the O3_offline simulation (a & d) and observations (b & e) are compared. The
 458 correlation coefficient (r), root-mean-square error (RMSE), normalized mean bias
 459 (NMB), and number of grid cells (n) for the comparisons are listed on the mean bias
 460 maps (c & f).

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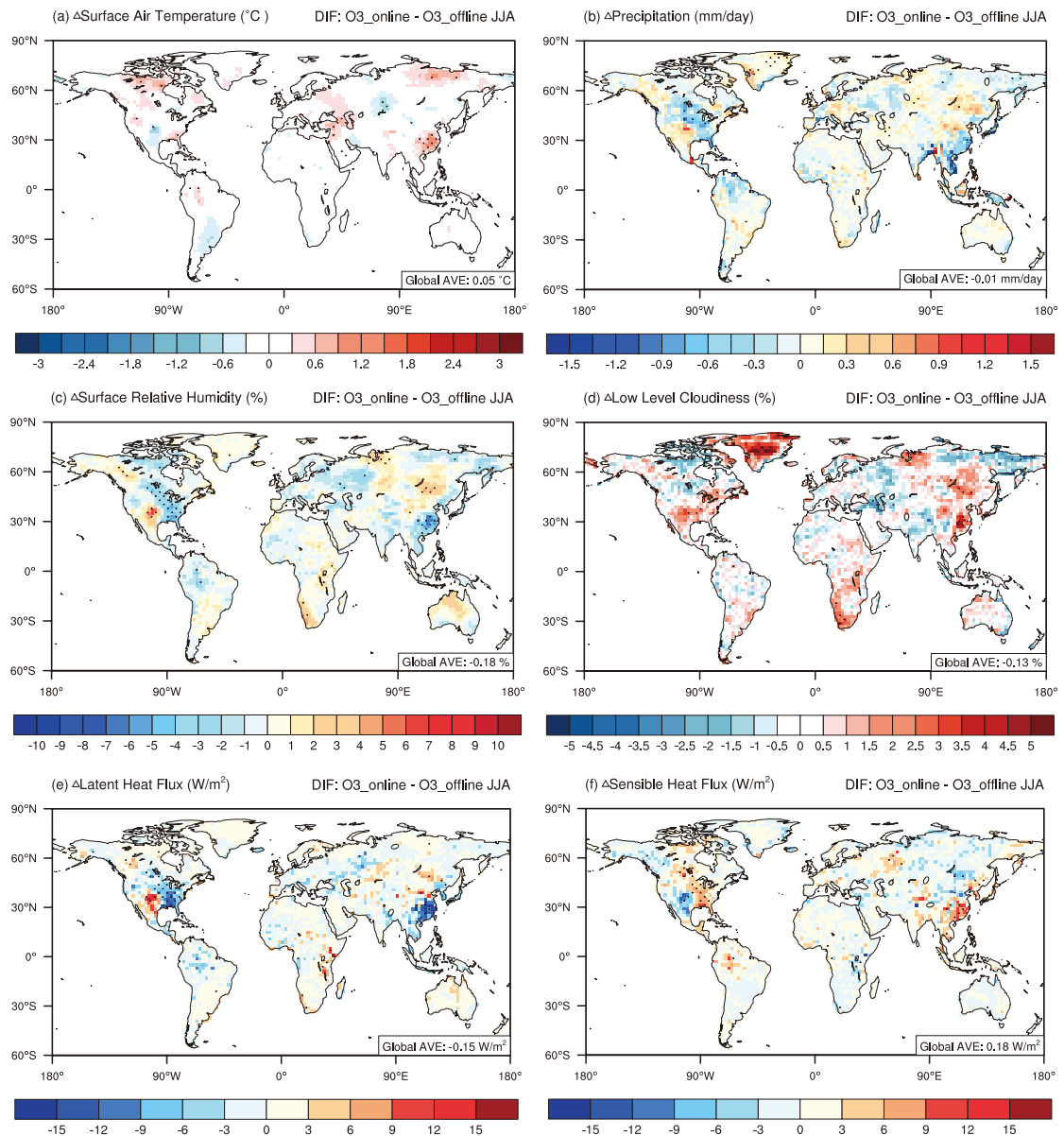
462

463 **Figure 2.** Same as in Fig. 1 but for gross primary productivity (GPP; a-c) and the leaf
464 area index (LAI; d-f).



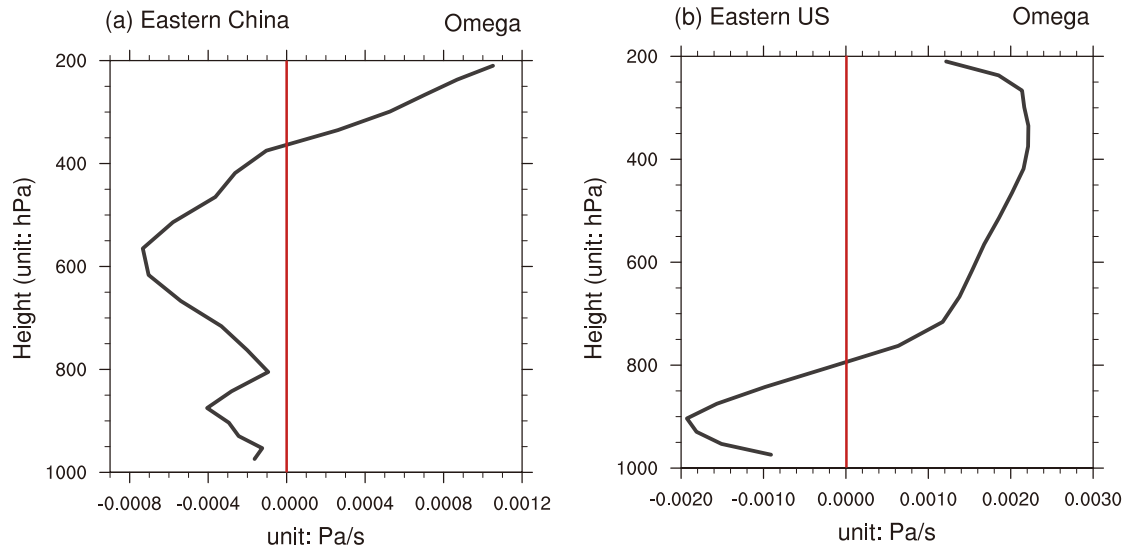
465

466 **Figure 3.** Changes in present-day boreal summertime biospheric variables induced by
 467 O₃-vegetation interactions. The results shown are the changes in (a) GPP, (b) canopy
 468 conductance, and (c) LAI between the O3_online and O3_offline simulations. The
 469 black dots denote areas with significant changes ($p < 0.1$).



470

471 **Figure 4.** Changes in present-day boreal summertime meteorological fields caused by
 472 O_3 -vegetation interactions. The results shown are changes in (a) surface air temperature,
 473 (b) precipitation, (c) surface relative humidity, (d) low-level cloudiness, (e) latent heat
 474 flux, and (f) sensible heat flux between the O_3_online and $O_3_offline$ simulations. For
 475 heat fluxes, positive values (shaded in red) indicate that the upward fluxes change. The
 476 black dots denote areas with significant changes ($p < 0.1$).



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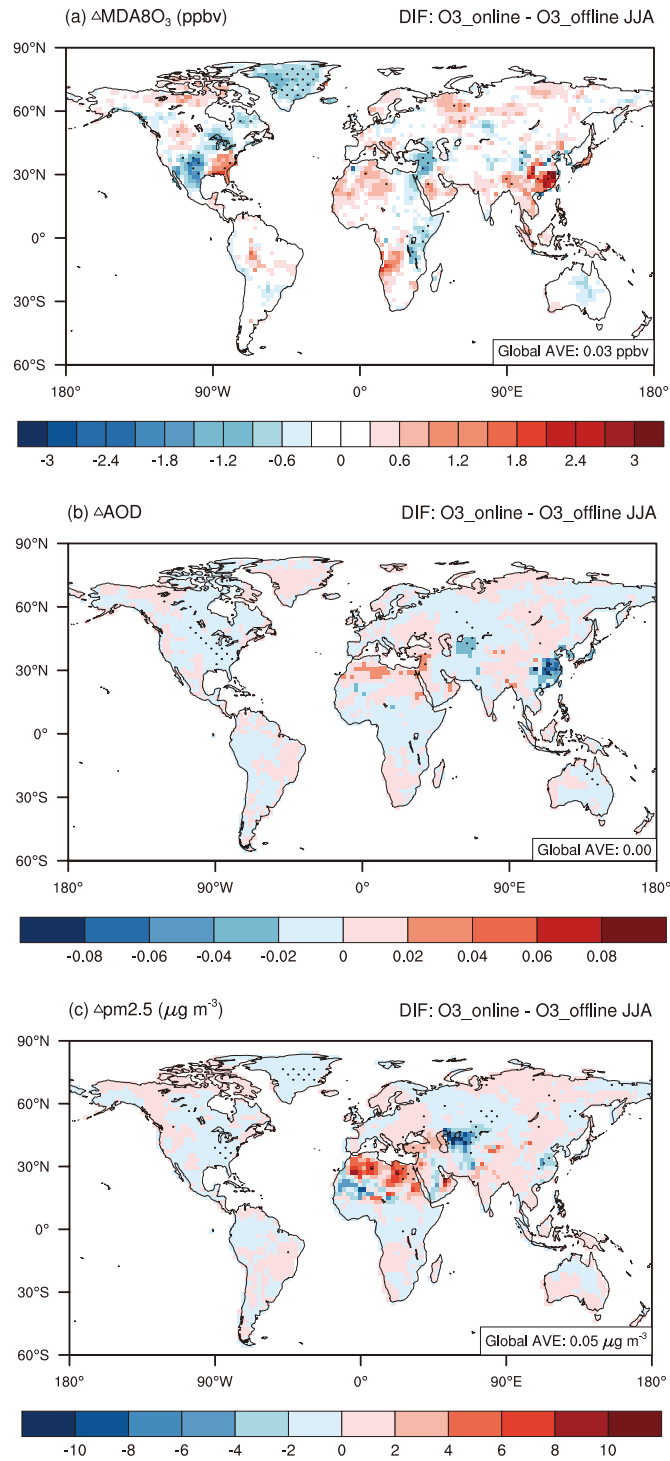
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Figure 5. Vertical profile of the vertical velocity. The results shown are changes in the vertical velocity in (a) eastern China and (b) the eastern U.S. between the O3_online and O3_offline simulations. The solid red line denotes 0. Please note the differences in the scales.



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Fig. 6. Changes in present-day summertime atmospheric pollution caused by O₃–vegetation interactions. The results shown are the changes in (a) O₃, (b) AOD, and (c) PM_{2.5} between the O3_online and O3_offline simulations. The black dots denote areas with significant changes ($p < 0.1$).

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