

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

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12 **Abstract.** The coupling between surface ozone (O<sub>3</sub>) and vegetation significantly  
13 influences regional to global climate. O<sub>3</sub> uptake by plant stomata inhibits  
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 Yale Interactive terrestrial Biosphere, or ModelE2-  
17 YIBs), we assess the global climatic responses to O<sub>3</sub>-vegetation interactions during  
18 boreal summer of the present day (2005-2014). High O<sub>3</sub> pollution reduces stomatal  
19 conductance, resulting in warmer and drier conditions worldwide. The most significant  
20 responses are found in the eastern U.S. and eastern China, where surface air temperature  
21 increases by  $+0.33\pm 0.87$  °C and  $+0.56\pm 0.38$  °C, respectively. These temperature rises  
22 are accompanied by decreased latent heat and increased sensible heat in both regions.  
23 The O<sub>3</sub>-vegetation interaction also affects atmospheric pollutants. Surface maximum  
24 daily 8-hour average O<sub>3</sub> concentrations increase by  $+1.46\pm 3.02$  ppbv in eastern China  
25 and  $+1.15\pm 1.77$  ppbv in eastern U.S due to the O<sub>3</sub>-induced inhibition of stomatal uptake.  
26 With reduced atmospheric stability following the warmer climate, increased cloudiness  
27 but decreased relative humidity jointly reduce aerosol optical depth by  $-0.06\pm 0.01$  (-  
28  $14.67\pm 12.15\%$ ) over eastern China. This study suggests that vegetation feedback should  
29 be considered for a more accurate assessment of climatic perturbations caused by  
30 tropospheric O<sub>3</sub>.

## 31 **1 Introduction**

32 Tropospheric ozone (O<sub>3</sub>), one of the most detrimental air pollutants (Myhre et al.,  
33 2013), not only poses threats to human health (Norval et al., 2011; Nuvolone et al., 2018)  
34 but also induces phytotoxic effects to vegetation (Mills et al., 2007; Pleijel et al., 2007).  
35 When exposed to certain levels of O<sub>3</sub>, plant photosynthesis and stomatal conductance  
36 is inhibited due to the O<sub>3</sub> oxidation of cellular, enzyme, and chlorophyll (Dizengremel,  
37 2001; Fiscus et al., 2005; Jolivet et al., 2016). Consequently, the carbon assimilation of  
38 terrestrial ecosystems is limited (Yue and Unger, 2014; Oliver et al., 2018) and the land-  
39 air exchange rates of water and heat fluxes are altered (Lombardozzi et al., 2015).

40 Experimental studies have shown that the excessive O<sub>3</sub> exposure reduced both  
41 plant photosynthesis and stomatal conductance (Ainsworth et al., 2012; Lombardozzi  
42 et al., 2013). The reduction rates are dependent on the O<sub>3</sub> stomatal fluxes as well as the  
43 damaging sensitivities that vary among different vegetation types (Nussbaum and  
44 Fuhrer, 2000; Karlsson et al., 2004; Pleijel et al., 2004). Several exposure-based indexes  
45 such as accumulated hourly O<sub>3</sub> concentrations over a threshold of 40 ppb (AOT40) and  
46 sum of all hourly average concentrations (SUM00) are used to assess O<sub>3</sub>-induced  
47 vegetation damage (Fuhrer et al., 1997; Paoletti et al., 2007). In addition, the flux-  
48 related POD<sub>y</sub> method (phytotoxic O<sub>3</sub> dose above a threshold flux of y) is also widely  
49 applied to consider the dynamic adjustment of stomatal conductance (Buker et al., 2015;  
50 Sicard et al., 2016). Taking into account the variability of plant sensitivities, different  
51 O<sub>3</sub> damage schemes were proposed to quantify the O<sub>3</sub> impacts on land carbon  
52 assimilation from regional to global scales (Anav et al., 2011; Lam et al., 2023; Lei et  
53 al., 2020). For example, Sitch et al. (2007) calculated the simultaneous damages to both  
54 photosynthesis and stomatal conductance based on the instantaneous O<sub>3</sub> stomatal  
55 uptake. In contrast, Lombardozzi et al. (2012) estimated the decoupled reductions in  
56 plant photosynthesis and stomatal conductance using different response relationships  
57 to the cumulative O<sub>3</sub> stomatal uptake. Applications of different schemes resulted in a  
58 wide range of reductions in gross primary productivity (GPP) by 2-12% globally with  
59 regional hotspots up to 20-30% (Lombardozzi et al., 2015; Unger et al., 2020; Zhou et  
60 al., 2024).

61 The O<sub>3</sub>-induced inhibition in stomatal conductance decreases dry deposition and  
62 consequently enhances surface O<sub>3</sub> concentrations (Clifton et al., 2020; Wesely and  
63 Hicks, 2000; Zhang et al., 2006). Using the Sitch et al. (2007) scheme with high O<sub>3</sub>  
64 damaging sensitivities in the ModelE2-YIBs (NASA GISS ModelE2 coupled with Yale  
65 Interactive terrestrial Biosphere model), Gong et al. (2020) revealed that O<sub>3</sub>-vegetation  
66 interactions increased regional O<sub>3</sub> concentrations by 1.8 ppbv in the eastern U.S., 1.3  
67 ppbv in Europe, and 2.1ppbv in eastern China for the year 2010. As a comparison, Sadiq  
68 et al. (2017) found consistently stronger feedback on O<sub>3</sub> concentrations in these polluted  
69 regions using the scheme of Lombardozzi et al (2012) embedded in the Community  
70 Earth System Model (CESM). Moreover, the inclusion of online O<sub>3</sub>-vegetation  
71 interactions in numerical models will also result in a greater loss of simulated land  
72 carbon assimilation due to the feedbacks of both ecosystems and surface O<sub>3</sub>. This is  
73 attributable to several factors. On one hand, O<sub>3</sub> damages to leaf photosynthesis inhibit  
74 plant growth and decrease leaf area index (LAI), leading to higher reduction percentage  
75 in GPP compared to simulations without LAI changes (Yue et al., 2020). On the other  
76 hand, the O<sub>3</sub> enhancement due to vegetation feedback may cause additional vegetation  
77 damage and result in further GPP losses (Lei et al., 2021). As a result, the O<sub>3</sub>-vegetation  
78 interactions should be considered in the global estimate of O<sub>3</sub> damages to ecosystem  
79 functions.

80 In addition to affecting surface O<sub>3</sub>, the O<sub>3</sub>-vegetation interaction can also alter the  
81 water and energy exchange between 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 O<sub>3</sub> by the leaves  
84 resulted in reduction of 2.2% in transpiration but increase of 5.4% in runoff globally.  
85 Arnold et al. (2018) used CESM and found that plant exposure to O<sub>3</sub> could decrease the  
86 land-air moisture fluxes and atmospheric humidity, which further reduced shortwave  
87 cloud forcing in polluted regions and induced widespread surface warming up to +1.5  
88 K. Two recent studies utilized the WRF-chem model and revealed considerable  
89 warming and the associated meteorological perturbations due to the O<sub>3</sub>-vegetation  
90 interactions in China (Zhu et al., 2022; Jin et al., 2023). However, all these modeling

91 studies applied the same O<sub>3</sub> vegetation damage scheme proposed by Lombardozzi et al.  
92 (2012). It's necessary to assess the climatic responses to O<sub>3</sub>-vegetation interactions  
93 using different schemes so as to explore the robust responses and the associated  
94 uncertainties.

95 In this study, we quantified the global impacts of O<sub>3</sub>-vegetation interaction on  
96 climatic conditions and surface air pollutants during 2010s using the ModelE2-YIBs  
97 (Yue and Unger, 2015). This fully coupled framework was implemented with the semi-  
98 mechanistic O<sub>3</sub> damage scheme proposed by Sitch et al. (2007), which calculated  
99 aggregated O<sub>3</sub> damage to photosynthesis based on varied sensitivities to instantaneous  
100 stomatal O<sub>3</sub> uptake across eight plant functional types (PFTs). We performed sensitivity  
101 experiments to quantify the responses of surface air temperature and precipitation to  
102 O<sub>3</sub>-vegetation interaction. The feedbacks to aerosols and O<sub>3</sub> concentrations were also  
103 examined.

104

## 105 **2 Method**

### 106 **2.1 Model descriptions**

107 The ModelE2-YIBs is a fully coupled climate-carbon-chemistry model combining  
108 the NASA GISS ModelE2 with the YIBs vegetation model. ModelE2 is a general  
109 circulation model with the horizontal resolution of 2°×2.5° in latitude and longitude  
110 and 40 vertical layers up to 0.1 hPa. It dynamically simulates gas-phase chemistry (NO<sub>x</sub>  
111 - HO<sub>x</sub> - O<sub>x</sub> - CO - CH<sub>4</sub> - NMVOCs), aerosols (sulfate, nitrate, black and organic carbon,  
112 dust, and sea salt), and their interactions (Menon and Rotstayn, 2006). Both the physical  
113 and chemical processes are calculated every 0.5 h and the radiation module is called  
114 every 2.5 h. The radiation module includes direct and indirect aerosol radiative effects  
115 and accounts for absorption of multiple greenhouse gases (GHGs). For cloud optical  
116 parameters, it uses Mie scattering, ray tracing, and matrix theory (Schmidt et al., 2006).  
117 The model outperforms 20 other IPCC-class climate models in simulating surface solar  
118 radiation (Wild et al., 2013) and has been extensively validated for meteorological and  
119 hydrological variables against observations and reanalysis data (Schmidt et al., 2014).

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

121 photosynthesis and Ball-Berry model for stomatal conductance (Farquhar et al., 1980;  
 122 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). The  $A_{tot}$   
 125 is derived from the minimum value of the constraints. The ribulose-1,5-bisphosphate  
 126 carboxylase (Rubisco) limited rate of carboxylation 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 rate for  $C_3$  plants and the phosphoenolpyruvate carboxylase (PEPC)  
 129 limited rate 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  $\text{CO}_2$  and oxygen partial pressure.  $\Gamma_*$  (Pa)  
 132 denotes the  $\text{CO}_2$  compensation point, while  $K_c$  and  $K_o$  (Pa) are Michaelis–Menten  
 133 constants for the carboxylation and oxygenation of Rubisco, respectively. The  
 134 parameters  $\Gamma_*$ ,  $K_c$ , and  $K_o$  vary with temperature based on 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 leaf-specific light absorbance that  
 137 considers sunlit and shaded leaves, and  $\alpha$  is quantum efficiency.  $P_{atm}$  (Pa) represents  
 138 the ambient pressure.  $K_s$  is set to 4000 as a constant following Oleson et al. (2010), to  
 139 limit photosynthesis of  $C_4$  plants get saturated at lower  $\text{CO}_2$  concentrations.

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

140 The stomatal conductance ( $g_s$ ,  $\text{mol} [\text{H}_2\text{O}] \text{m}^{-2} \text{s}^{-1}$ ) is linked to the variations of  $A_{tot}$   
 141 with parameters such as dark respiration rate ( $R_d$ ,  $\mu\text{mol m}^{-2} \text{s}^{-1}$ ), relative humidity ( $RH$ ),

142 and CO<sub>2</sub> concentration at the leaf surface ( $c_s$ ). The model simulates the biophysical  
 143 processes of eight PFTs including tundra, C<sub>3</sub>/C<sub>4</sub> grass, shrubland, deciduous broadleaf  
 144 forest, evergreen broadleaf forest, evergreen needleleaf forest, and cropland. Different  
 145 values are assigned to parameters  $m$  and  $b$  for each PFT (Table S1). The carbon uptake  
 146 by the leaf is then accumulated and allocated to different organs to support the plant  
 147 development with dynamical changes in LAI and tree growth.

148

## 149 **2.2 The O<sub>3</sub>-vegetation damage scheme**

150 The YIBs model employs a semi-mechanistic parameterization proposed by Sitch  
 151 et al. (2007) to estimate the impact of O<sub>3</sub> on photosynthesis through stomatal uptake.  
 152 The scheme applies an undamaged factor ( $F$ ) (nmol m<sup>-2</sup> s<sup>-1</sup>) to both  $A_{tot}$  and  $g_s$  as  
 153 follows:

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

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

154 where  $A_{totd}$  and  $g_{sd}$  are the unaffected photosynthesis and stomatal conductance  
 155 separately. The factor  $F$  is defined as:

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

156  $a_h$  (mmol m<sup>-2</sup> s<sup>-1</sup>) is the high O<sub>3</sub> sensitivity coefficient, calibrated by Sitch et al. (2007)  
 157 on data from field observations by Karlsson et al. (2004) and Pleijel et al. (2004) to  
 158 represent ‘high’ sensitivity of relative species of each PFT.  $F_{O_3,crit}$  (nmol m<sup>-2</sup> s<sup>-1</sup>) is the  
 159 specific threshold for O<sub>3</sub> damages, both of which varies with vegetation types (Table  
 160 S1).

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

161 where  $[O_3]$  represents surface O<sub>3</sub> concentrations,  $R_a$  (s m<sup>-1</sup>) stands for aerodynamic  
 162 resistance, which expresses turbulent transport efficiency in transferring sensible heat  
 163 and water vapor between the land surface and a reference height. The constant  
 164  $k_{O_3}=1.67$  is the ratio of stomatal resistance for O<sub>3</sub>, estimated based on the theoretical  
 165 stomatal resistance to water (Laisk et al., 1989). When plants are exposed to  $[O_3]$  (Eq.  
 166 9),  $A_{tot}$  and  $g_s$  will decrease (Eq. 6 and Eq. 7) if the excess O<sub>3</sub> enters leaves (Eq. 8).

167 The increased stomatal resistance acts to protect plants by reducing the O<sub>3</sub> uptake of  
168 stomata. Consequently, the damage scheme describes both changes in photosynthetic  
169 rate and stomatal conductance.

170

### 171 **2.3 Experiments**

172 To explore the coupled O<sub>3</sub>-vegetation effect, we performed two simulations using  
173 the ModelE2-YIBs model. The control experiment “O<sub>3</sub>\_offline” was conducted  
174 without the O<sub>3</sub> damages to vegetation. As a comparison, the sensitivity experiment  
175 “O<sub>3</sub>\_online” contained online O<sub>3</sub>-vegetation interaction with high O<sub>3</sub> sensitivity. For  
176 both experiments, the anthropogenic emissions of 2010 (the average of 2005-2014) for  
177 8 species (BC, OC, CO, NH<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub>, Alkenes, and Paraffin) from 8 economic  
178 sources (agriculture, energy, industry, transportation, resident, solvent, waste, and  
179 international shipping) and biomass burning source were collected from the Coupled  
180 Model Intercomparison Project phase 6 (CMIP6) (van Marle et al., 2017; Hoesly et al.,  
181 2018). The ensemble mean of monthly sea surface temperature (SST) and sea ice  
182 fraction (SIC) simulated by 21 CMIP6 models during the time period 2005-2014 was  
183 employed as the boundary conditions. The cover fraction of 8 PFTs (Fig. S1) fixed at  
184 2010 were adopted from the land use harmonization (LUH2) dataset (Hurtt et al., 2020).  
185 For each time-slice simulation, the model was run for 30 years with all the input data  
186 fixed and the first 10 years are used as the spin up. We calculated the average of the last  
187 20 years and focused on the boreal summer season (June-July-August, JJA) when the  
188 interaction of vegetation and surface O<sub>3</sub> reaches the maximum in one year (fig. S3). In  
189 order to show the uncertainty introduced by the internal variability of the model, all the  
190 related global/regional values are denoted as “mean/sum ± standard deviation of the  
191 last 20 model years”. We explored the climatic responses to O<sub>3</sub>-vegetation interactions  
192 as the differences between “O<sub>3</sub>\_online” and “O<sub>3</sub>\_offline” on the global scale with the  
193 focus over the hotspot regions such as eastern U.S. (30–40° N, 80–90° W) and eastern  
194 China (22.5–38° N, 106–122° E).

195

### 196 **2.4 Data for model evaluation**



197 We evaluated the simulated air pollutants, carbon fluxes, and meteorological  
198 variables from ‘O3\_offline’ run using observational and reanalysis datasets. The  
199 worldwide observations of the maximum daily 8-hour average O<sub>3</sub> (MDA8 O<sub>3</sub>)  
200 concentrations were mainly collected from three regional networks: Air Quality  
201 Monitoring Network operated by Ministry of Ecology and Environment (AQMN-MEE)  
202 in China, the Clean Air Status and Trends Network (CASTNET) in the U.S., and the  
203 European Monitoring and Evaluation Programme (EMEP) in Europe. Observations  
204 used for validation beyond China, sourced from Sofen et al. (2016), are averaged over  
205 the period 2005-2014. This dataset encompasses 7288 station records worldwide and  
206 excludes the uncertainty associated with high mountain-top sites. For AQMN-MEE, the  
207 mean value of 2014-2018 was used due to its establishment in 2013. The simulated  
208 aerosol optical depth (AOD) and LAI were validated using satellite-based data from the  
209 Moderate Resolution Imaging Spectroradiometer (MODIS) retrievals collection 5  
210 (Remer et al., 2005) (<http://modis.gsfc.nasa.gov/>) averaged for the years 2005-2014.  
211 The simulated GPP was evaluated against the data product upscaled from the  
212 FLUXNET eddy covariance measurements for 2009-2011 (Jung et al., 2011). The daily  
213 temperature at 2m (T<sub>2m</sub>) in 2005-2014 was obtained from the National Centers for  
214 Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR)  
215 reanalysis 1 (NCEP1) (Kalnay et al., 1996). For precipitation, we used the monthly data  
216 averaged in 2005-2014 from Global Precipitation Climatology Project (GPCP)  
217 (Huffman et al., 1997; Adler et al., 2018). All these datasets were interpolated to the  
218 same resolution as ModelE2-YIBs model. Root-mean-square-error (RMSE) and  
219 normalized mean biases (NMBs) were applied to quantify the deviations of simulations  
220 from observations:

$$221 \quad RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (S_i - O_i)^2} \quad (10)$$

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

222 Here, S<sub>i</sub> and O<sub>i</sub> represent the simulated and observed values, respectively. *n* denotes the  
223 total grid number used in the comparisons.

224

## 225 **3. Results**

### 226 **3.1 The control simulation and model evaluations**

227 We first evaluated the air pollutants simulated by the control simulation O3\_offline  
228 of ModelE2-YIBs model (Fig. 1). Over a total of 503 grids with site-level O<sub>3</sub>  
229 measurements (Fig. 1b), the model replicated both the magnitude and spatial  
230 distribution of MDA8 O<sub>3</sub>, with correlation coefficient ( $r$ ) of 0.59 and NMB of -2.54%  
231 (Fig. 1c). Simulated summertime surface MDA8 O<sub>3</sub> was high in regions with large  
232 anthropogenic emissions, such as western Europe and eastern China (Ohara et al., 2007),  
233 as well as in central Africa with frequent fire emissions (van der Werf et al., 2017). On  
234 the global scale, the model yielded an average MDA8 O<sub>3</sub> of 43.93 ppbv and  
235 observations showed an average of 44.72 ppbv over the same grids. However, the  
236 modeled result is overestimated over the North China Plain and slightly underestimated  
237 over the U.S., likely due to the biases in the emission inventories and predicted climate  
238 that drive the O<sub>3</sub> production. Simulated AOD at 550 nm by O3\_offline (Fig. 1d) showed  
239 similar spatial pattern as the satellite retrievals (Fig. 1e) with  $R=0.75$  and NMB of -  
240 7.35% globally (Fig. 1f). Both the simulations and observations showed AOD hotspots  
241 over North Africa and the Middle East where dust emissions dominate, and in northern  
242 India and eastern China where anthropogenic emissions are large (Feng et al., 2020).

243 We then evaluated the simulated GPP and LAI by the control experiment for the  
244 boreal summer period (Fig. 2). Observations showed GPP hotspots over boreal forests  
245 such as eastern U.S., Eurasia, and East Asia and the tropical forests such as Amazon,  
246 central Africa, and Indonesia (Fig. 2b). The seasonal total GPP was estimated to be  
247 41.63Pg[C], which accounted for 35% of the annual amount. Simulations captured the  
248 observed GPP pattern on the global scale, with  $r = 0.64$  and NMB = -7.81% over 2581  
249 grids (Fig. 2c), with underestimation in the tundra area and slight overestimation in the  
250 tropical rain forest and evergreen forest regions. The model simulated a seasonal total  
251 GPP of 38.69 Pg[C], equivalent to 34% of the annual amount. Simulated LAI showed  
252 similar patterns as GPP (Fig. 2d) and resembled observed LAI (Fig. 2e) with a spatial  
253 correlation  $r = 0.79$  and a low NMB = -5.43% over 4435 grids globally (Fig. 2f).

254 We further validated the simulated meteorology from O3\_offline (Fig. S2). For

255 surface air temperature, the model (Fig. S2a) reproduced observed (Fig. S2b) pattern  
256 with RMSE of 3.21 °C and r of 0.99 against observations (Fig. S2c). For precipitation,  
257 the simulation (Fig. S2d) captures the observed spatial pattern (Fig. S2e) with NMB =  
258 17.26% and r = 0.75 (Fig. S2f). Overall, the model captures the spatial characteristics  
259 and magnitudes of air pollutants, biospheric parameters, and meteorological fields,  
260 making it a valuable tool for studying O<sub>3</sub>-vegetation interactions.

261

### 262 **3.2 O<sub>3</sub> damage to terrestrial ecosystems**

263 We assessed the damaging effects of surface O<sub>3</sub> to ecosystems due to online O<sub>3</sub>-  
264 vegetation interactions (Fig. 3). The impacts of O<sub>3</sub> on biospheric variables were mainly  
265 located in regions characterized by abundant vegetation cover and elevated O<sub>3</sub>  
266 concentrations. On the global scale, O<sub>3</sub> induced the GPP reduction of  $-1.80 \pm 0.61$  PgC  
267 yr<sup>-1</sup> ( $-4.69 \pm 1.56\%$ , Fig. 3a). This deleterious effect was more pronounced in specific  
268 regions, notably eastern China and eastern U.S., with significant GPP declines of -  
269  $25.40 \pm 1.90\%$  and  $-20.14 \pm 5.02\%$ , respectively, under high O<sub>3</sub> sensitivity conditions (Fig.  
270 3a and Table S2). Meanwhile, stomatal conductance significantly decreased in the  
271 middle latitudes of Northern Hemisphere (Fig. 3b). The most substantial relative change  
272 of  $-30.62 \pm 4.30\%$  was observed in eastern China, followed by  $-25.65 \pm 9.32\%$  in the  
273 eastern U.S. (Fig. 3b and Table S2). Though there are positive responses in some  
274 regions, they are not dominant and hardly significant. These values were stronger than  
275 that for GPP (Fig. 3a), likely due to the climatic feedback to O<sub>3</sub>-vegetation interactions.  
276 The opening of plant stoma plays a crucial role in regulating the energy and water  
277 exchange between land surface and the atmosphere. The inhibition of stomatal  
278 conductance by surface O<sub>3</sub> leads to the warmer (Fig. 4a) and drier (Fig. 4b) climate in  
279 those hotspot regions, resulting in even stronger inhibition effects on stomatal  
280 conductance. Following the changes in GPP, global LAI on average decreased by  
281  $0.01 \pm 0.01$  m<sup>2</sup> m<sup>-2</sup> ( $-0.62 \pm 0.84\%$ ) with regional maximums of  $-4.53 \pm 1.14\%$  in eastern  
282 China and  $-5.87 \pm 3.11\%$  in eastern U.S. (Table S2).

283

### 284 **3.3 Global climatic responses to O<sub>3</sub>-vegetation interactions**

285 In response to the O<sub>3</sub>-induced inhibition of stomatal conductance, surface air  
286 temperature increased by  $0.05\pm 0.20^{\circ}\text{C}$  (Fig. 4a) while precipitation decreased by -  
287  $0.01\pm 0.03\text{ mm day}^{-1}$  (Fig. 4b) on the global scale. The most significant change was the  
288 warming of  $0.56\pm 0.38^{\circ}\text{C}$  and precipitation reduction of  $-0.79\pm 1.05\text{ mm day}^{-1}$  (-  
289  $16.18\pm 20.38\%$ ) in eastern China (Table S3), following the largest inhibition to stomatal  
290 conductance (Fig. 3b). Such warming and rainfall deficit also appeared in eastern U.S.  
291 and western Europe, where the O<sub>3</sub>-vegetation interactions were notable. The O<sub>3</sub>-  
292 induced inhibition to stomatal conductance decreased latent heat flux (Fig. 4e) and the  
293 consequent precipitation (Fig. 4b) in those hotspot regions. Meanwhile, the reduction  
294 of latent heat flux promotes surface air temperature (Fig. 4a), resulting in the increase  
295 of sensible heat flux (Fig. 4f). Such warming was also reported in field experiments,  
296 where relatively high O<sub>3</sub> exposure resulted in noticeable increases of canopy  
297 temperature along with reductions of transpiration (Bernacchi et al., 2011; VanLoocke  
298 et al., 2012). Globally, temperature and precipitation showed patchy responses with  
299 both positive and negative anomalies, suggesting that the regional hotspots of O<sub>3</sub>-  
300 induced meteorological changes propagate to surrounding areas through atmospheric  
301 perturbations.

302 We further examined the changes in air humidity and cloudiness. Surface relative  
303 humidity decreased by  $-0.18\pm 0.53\%$  globally with a similar pattern as that of  
304 precipitation (Fig. 4c). The most significant reductions were over eastern China and  
305 eastern U.S., where both the warming (Fig. 4a) and rainfall deficit (Fig. 4b) contributed  
306 to the drought. However, in the adjacent regions such as northern China and central  
307 U.S., both rainfall and surface relative humidity showed certain enhancement. These  
308 changes were associated with the regional increase of cloud cover (Fig. 4d). The  
309 sensible heat flux increased by  $6.3\pm 5.4\text{ W m}^{-2}$  ( $16.54\pm 15.59\%$ ) and  $7.12\pm 3.86\text{ W m}^{-2}$   
310 ( $25.46\pm 14.71\%$ ) in eastern U.S. and eastern China, respectively, suggesting a transfer  
311 of thermal energy from land to the atmosphere by O<sub>3</sub>-vegetation interactions (Fig. 4f  
312 and Table S3). The warming effect further triggered anomalous updrafts in the lower  
313 troposphere, represented by the changes in vertical velocity (Fig. 5), leading to

314 enhanced convection, reduced atmospheric stability, and consequently an increase in  
315 low-level cloudiness (Fig. 4d). However, despite the usual cooling effect associated  
316 with increased cloud cover due to reductions in radiation, in regions predominantly  
317 influenced by O<sub>3</sub>-vegetation interactions, this cooling effect was outweighed by the O<sub>3</sub>-  
318 induced warming through inhibition of stomatal conductance. Therefore, temperatures  
319 exhibited an overall increase of 0.56±0.38°C in eastern China and 0.33±0.87 °C in the  
320 eastern U.S. (Table S3).

321

### 322 **3.4 Changes of air pollution by O<sub>3</sub>-vegetation interactions**

323 Changes in surface water and heat fluxes induced by O<sub>3</sub>-vegetation interactions  
324 could feed back to affect air pollutants such as O<sub>3</sub> and aerosols. As Fig. 6a and Table  
325 S4 show, surface MDA8 O<sub>3</sub> concentrations enhanced 1.46±3.02 ppbv in eastern China  
326 and 1.15±1.77 ppbv in eastern U.S. due to the decreased dry deposition following O<sub>3</sub>  
327 inhibition on stomatal conductance. It indicates that the high contemporary O<sub>3</sub> pollution  
328 may worsen air quality through O<sub>3</sub>-vegetation interactions. However, negative O<sub>3</sub>  
329 changes were predicted in central U.S. and western China, where the increased rainfall  
330 dampened O<sub>3</sub> through chemical reactions and wet deposition. On a global scale, surface  
331 MDA8 O<sub>3</sub> showed a limited increase of 0.03±0.4 ppbv due to the offset between  
332 positive and negative feedbacks. The enhancement of O<sub>3</sub> concentrations in polluted  
333 regions may exacerbate the warming effect of O<sub>3</sub> as a greenhouse gas and cause  
334 additional damages to vegetation. For instance, offline O<sub>3</sub> damages on GPP in eastern  
335 China and the eastern US are -0.52±0.03 Pg[C] (-24.98±0.91%) and -0.17±0.02 Pg[C]  
336 (-16.71±1.16%), respectively, smaller than those induced by O<sub>3</sub>-vegetation interactions  
337 (Table S2).

338 Aerosols also exhibited evident changes by the O<sub>3</sub>-vegetation interactions. The  
339 AOD showed significant reductions over the hotspot regions such as eastern China and  
340 eastern U.S. (Fig. 6b). In the ModelE2-YIBs model, sulfate was especially sensitive to  
341 cloud which could enhance the aerosol scavenging through cloud water precipitation  
342 (Koch et al., 2006). The large enhancement of cloudiness removed sulfate more  
343 efficiently than other aerosol species, leading to an average decline of -1.94±1.67 μg

344  $\text{m}^{-3}$  ( $-8.52\pm 6.88\%$ ) in  $\text{PM}_{2.5}$  loading over eastern China (Fig. S4 and Table S4).  
345 Meanwhile, the reduction of surface relative humidity (Fig. 4c) in the regions with  
346 strong  $\text{O}_3$ -vegetation interactions limited the hygroscopic growth of aerosols, leading  
347 to a more noticeable decrease in AOD (Petters and Kreidenweis, 2007; Pitchford et al.,  
348 2007) by  $-0.06\pm 0.05$  ( $-14.67\pm 16.75\%$ ) in eastern China (Table S4). The similar aerosol  
349 changes were found in eastern U.S. but with smaller reductions of  $\text{PM}_{2.5}$  by  $-0.27\pm 0.36$   
350  $\mu\text{g m}^{-3}$  ( $-6.01\pm 7.9\%$ ) and AOD by  $-0.01\pm 0.01$  ( $-8.15\pm 9.38\%$ ) (Table S4). Beyond the  
351 key  $\text{O}_3$ -vegetation coupling regions, positive but insignificant changes in AOD were  
352 predicted, leading to the moderate AOD changes on the global scale (Fig. 6b).

353

#### 354 **4. Discussion and conclusions**

355 We examined the  $\text{O}_3$ -vegetation feedback to climate and air pollution in the 2010s  
356 using the fully coupled climate-carbon-chemistry model ModelE2-YIBs. During boreal  
357 summer, surface  $\text{O}_3$  resulted in strong damages to GPP and inhibitions to stomatal  
358 conductance with regional hotspots over eastern China and eastern U.S. Consequently,  
359 surface transpiration was weakened, leading to decreased latent heat fluxes and relative  
360 humidity but increased surface air temperature. Meanwhile, the surface warming  
361 increased cloud cover by reducing atmospheric stability. However, the enhancement of  
362 cloudiness decreased surface temperature and promoted precipitation outside the key  
363 regions with intense  $\text{O}_3$ -vegetation interactions. The  $\text{O}_3$ -induced inhibition to stomatal  
364 conductance resulted in a localized increase in  $\text{O}_3$  concentrations. In contrast, the  
365 increased cloud cover and decreased relative humidity jointly reduced AOD in hotspot  
366 regions. On the global scale, the mean changes of both climate and air pollution were  
367 moderate due to the offset between the changes with opposite signs.

368 Our predicted changes in water/heat fluxes by  $\text{O}_3$ -vegetation interactions were  
369 consistent with previous studies (Lombardozzi et al., 2015; Arnold et al., 2018; Gong  
370 et al., 2020). For example, the simulations by Lombardozzi et al. (2015) revealed that  
371 surface  $\text{O}_3$  reduces global GPP by 8%-12% and transpiration by 2-2.4% with regional  
372 reductions up to 20% for GPP and 15% for transpiration in eastern China and U.S.  
373 These changes were in general consistent with our results though we predicted larger

374 reductions in transpiration than GPP due to O<sub>3</sub>-vegetation interactions. Using the same  
375 scheme as Lombardozzi et al. (2015), Sadiq et al. (2017) showed that O<sub>3</sub>-vegetation  
376 coupling induced the surface warming of 0.5-1°C and O<sub>3</sub> enhancement of 4-6 ppbv in  
377 eastern China and eastern U.S. The magnitude of these responses was much stronger  
378 than our predictions, likely because they considered the accumulation effect of O<sub>3</sub>. In  
379 contrast, the regional simulations by Jin et al. (2023) revealed that O<sub>3</sub>-vegetation  
380 coupling led to the increases of temperature up to 0.16°C and surface O<sub>3</sub> up to 0.6 ppbv  
381 in eastern China, both of which were smaller than our predictions. The damage scheme  
382 they use, which depends on cumulative O<sub>3</sub> uptake, omits the difference in impact on  
383 sunlit or shaded leaves and will overestimate the O<sub>3</sub> damage on GPP compared to the  
384 scheme we use, which considers transient O<sub>3</sub> flux (Cao et al., 2024). The discrepancies  
385 of O<sub>3</sub>-vegetation feedback using the same O<sub>3</sub> damage schemes revealed the  
386 uncertainties from climate and chemistry models. Our predictions were within the range  
387 of previous estimates for both climatic and O<sub>3</sub> changes.

388         There were some limitations in our simulated O<sub>3</sub>-vegetation interactions. First, the  
389 semi-mechanistic O<sub>3</sub> damage scheme we used in the study linked the damages to  
390 photosynthesis with those to stomatal conductance (Sitch et al., 2007), leading to  
391 stronger inhibition percentage in stomatal conductance than that in photosynthesis  
392 considering the O<sub>3</sub>-vegetation feedback. However, some observations showed that the  
393 damage to stomatal conductance occurred more slowly and might not be proportional  
394 to the decline of photosynthetic rates (Gregg et al., 2006; Lombardozzi et al., 2012).  
395 Second, observations have shown large variability of plant sensitivities to O<sub>3</sub> damages.  
396 The Sitch et al. (2007) scheme employed the low to high ranges of sensitivity to indicate  
397 the inter-specific variabilities. In this study, we employed only the high O<sub>3</sub> sensitivity  
398 to explore the maximum responses. The possible uncertainties due to varied O<sub>3</sub> damage  
399 sensitivities deserved further investigations. Third, large-scale observations were not  
400 available to validate the simulated regional to global responses of climate and air  
401 pollutants. The O<sub>3</sub> vegetation damage scheme was extensively validated against site-  
402 level measurements of both photosynthesis (Yue and Unger, 2018) and stomatal

403 conductance (Yue et al., 2016). However, we were conservative about the derived  
404 global responses given that previous studies showed large discrepancies using the same  
405 O<sub>3</sub> damage scheme but implemented in different climate and/or chemistry models  
406 ( Lombardozzi et al., 2015; Sadiq et al., 2017; Jin et al., 2023). Furthermore, the 2°×2.5°  
407 resolution of current ModelE2-YIBs has limitation due to the high computational  
408 demands. However, high-resolution models exhibit improved simulations of extreme  
409 events (Chang et al., 2020; Ban et al., 2021), which have certain effect on O<sub>3</sub>-vegetation  
410 interactions (Mills et al., 2016; Lin et al., 2020). While chemical transport models with  
411 relatively coarse resolution can raise biases in simulated air pollutants, they still capture  
412 large-scale patterns similar to fine-resolution results and is reasonable compared to  
413 observational data (Wang et al., 2013; Li et al., 2016; Lei et al., 2020). Moreover, we  
414 omit the slow climatic feedback caused by air-sea interaction in the simulations. Studies  
415 have revealed that these interactions may result in different climatic perturbations from  
416 those simulations with fast responses of land surface alone (Yue et al., 2011). A dynamic  
417 ocean model is considered to enrich the future research. Meanwhile, this study does not  
418 isolate the different impacts of aerosols, even though the radiation module includes both  
419 direct and indirect radiative effects. We will investigate this further in the future by  
420 identifying the main processes.

421 Despite these uncertainties, our simulations revealed considerable changes of both  
422 climate and air pollutants in response to O<sub>3</sub>-vegetation interactions. The most intense  
423 warming, dryness, and O<sub>3</sub> enhancement were predicted in eastern China and eastern  
424 U.S., affecting the regional climate and threatening public health for these top two  
425 economic centers. In contrast, we for the first time revealed the reduction of aerosol  
426 loading in those hotspot regions, suggesting both positive and negative effects to air  
427 pollutants by O<sub>3</sub>-vegetation feedback. Such interactions should be considered in the  
428 Earth system models so as to better project future changes in climate and air pollutants  
429 following the anthropogenic interventions to both O<sub>3</sub> precursor emissions and  
430 ecosystem functions.

431



432 **Data Availability**

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

435

436 **Author contributions**

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

440

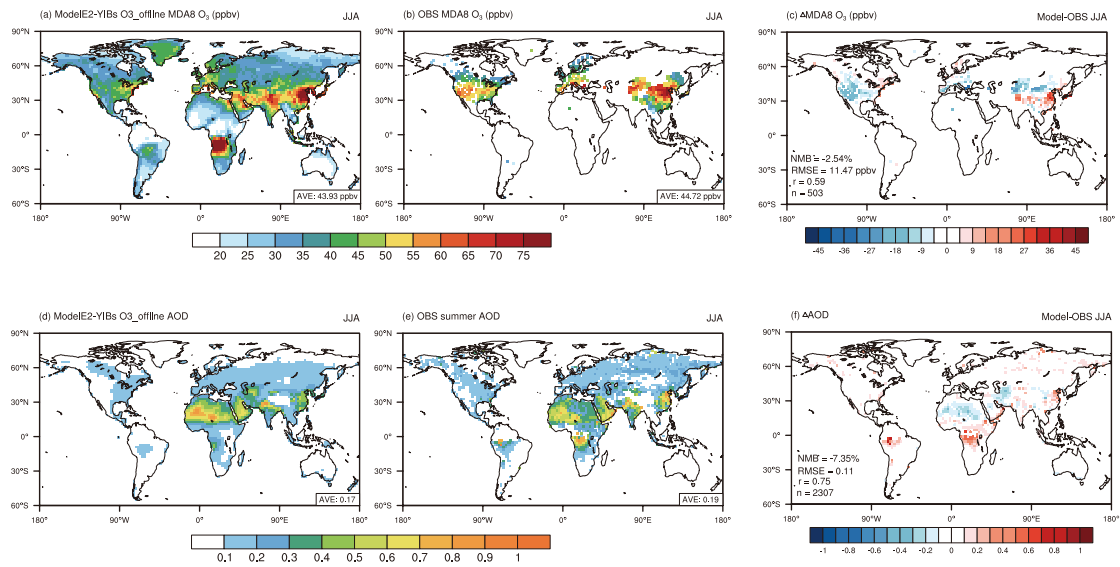
441 **Competing interests**

442 The authors declare that they have no conflict of interest.

443

444 **Acknowledgments**

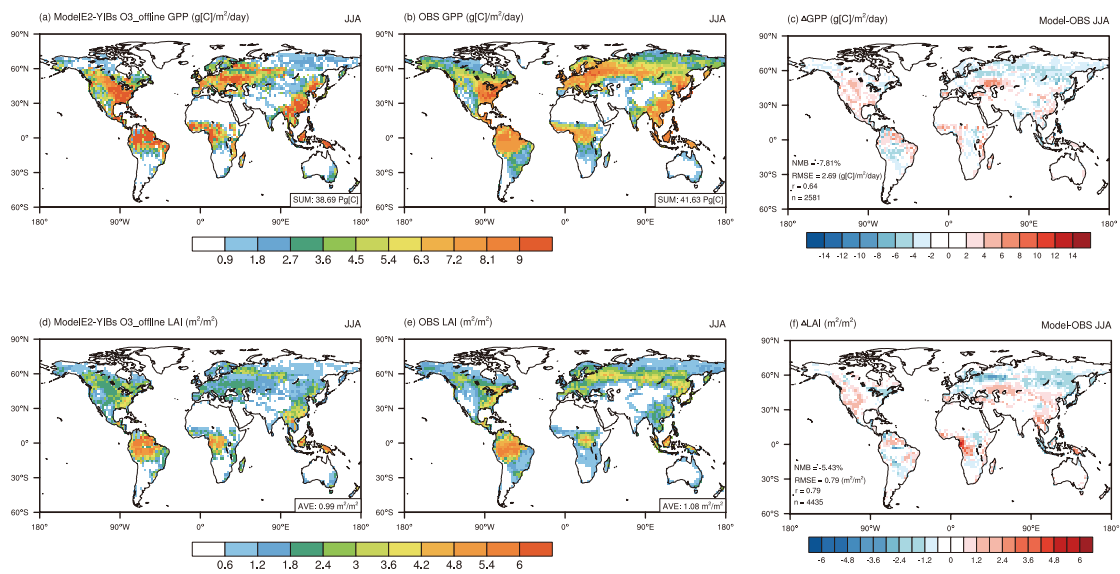
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447 42293323).



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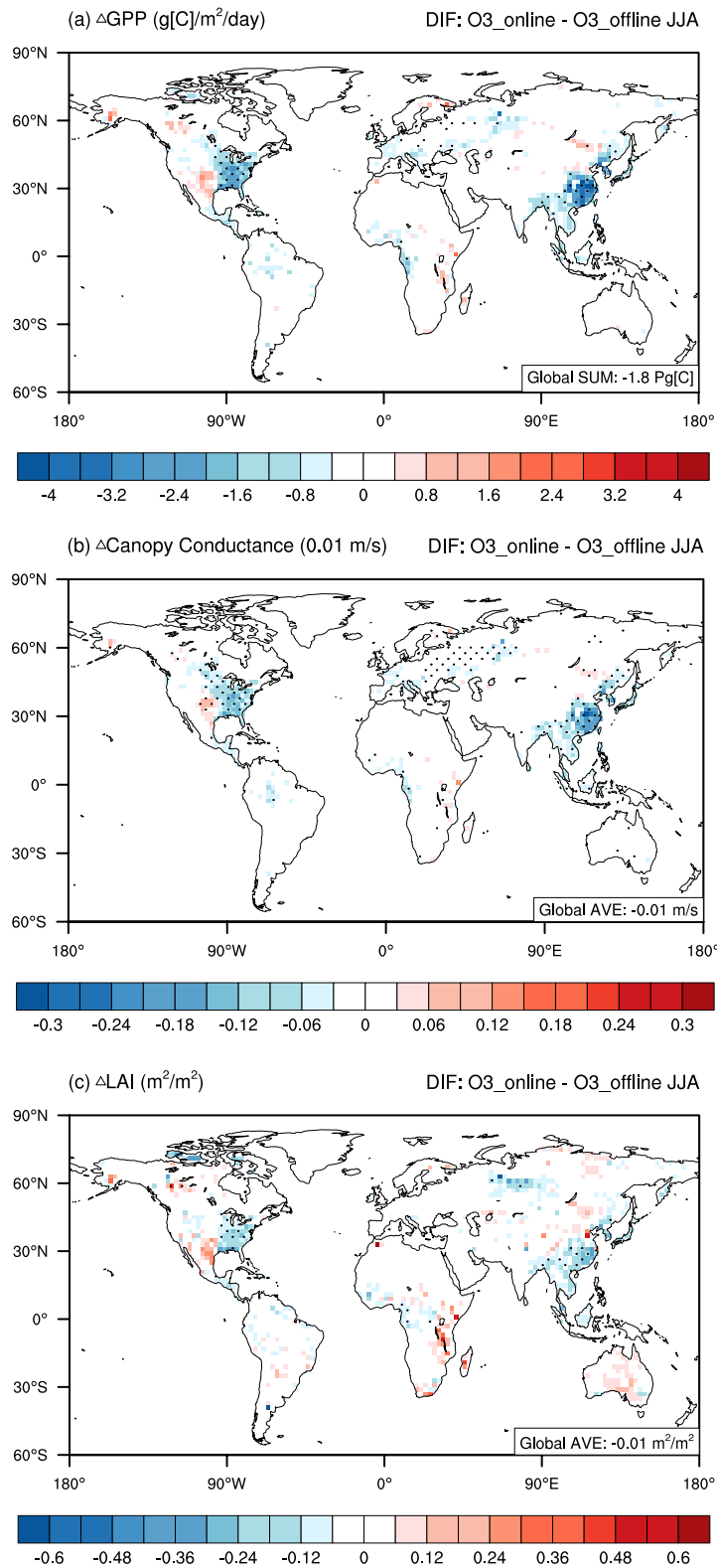
449 **Figure 1.** Evaluation of the boreal summertime (June-August) air pollutants at the  
 450 present day simulated by the ModelE2-YIBs model. Surface daily maximum 8-hour  
 451 ozone (MDA8 O<sub>3</sub>; a-c) and aerosol optical depth (AOD; d-f) from the simulation  
 452 O3\_offline (a & d) and observations (b & e) are compared. The correlation coefficients  
 453 ( $r$ ), root mean square error (RMSE), normalized mean bias (NMB), and number of grid  
 454 cells ( $n$ ) for the comparisons are listed on the mean bias maps (c & f).

455



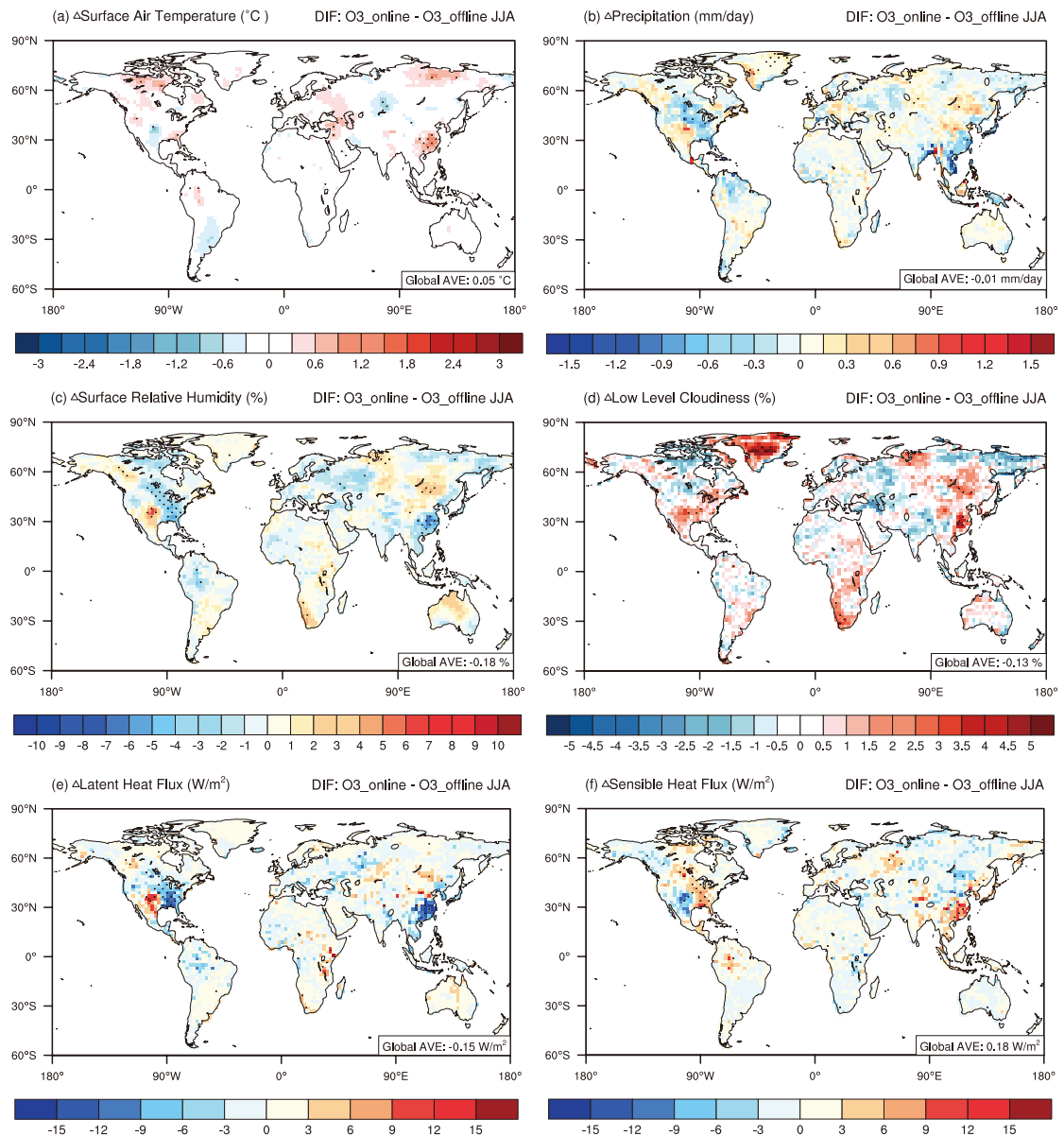
456

457 **Figure 2.** The same as Fig.1 but for gross primary productivity (GPP; a-c) and leaf area  
458 index (LAI; d-f).



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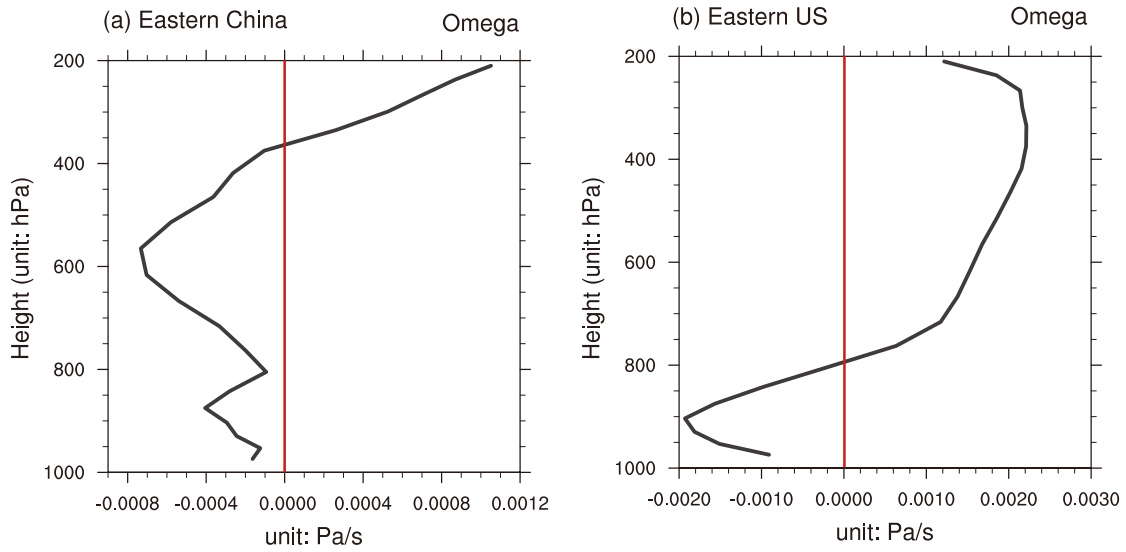
460 **Figure 3.** Changes of boreal summertime biospheric variables induced by O<sub>3</sub>-  
 461 vegetation interactions at the present day. Results shown are changes of (a) GPP, (b)  
 462 canopy conductance, and (c) LAI between simulations O3\_online and O3\_offline.  
 463 Black dots denote areas with significant changes ( $p < 0.1$ ).



464

465 **Figure 4.** Changes of boreal summertime meteorological fields by O<sub>3</sub>-vegetation  
 466 interactions at the present day. Results shown are changes of (a) surface air temperature,  
 467 (b) precipitation, (c) surface relative humidity, (d) low level cloudiness, (e) latent heat  
 468 flux, and (f) sensible heat flux between simulations O<sub>3</sub>\_online and O<sub>3</sub>\_offline. For heat  
 469 fluxes, positive values (shaded in red color) indicate the upward fluxes change. Black  
 470 dots denote areas with significant changes ( $p < 0.1$ ).

471



472

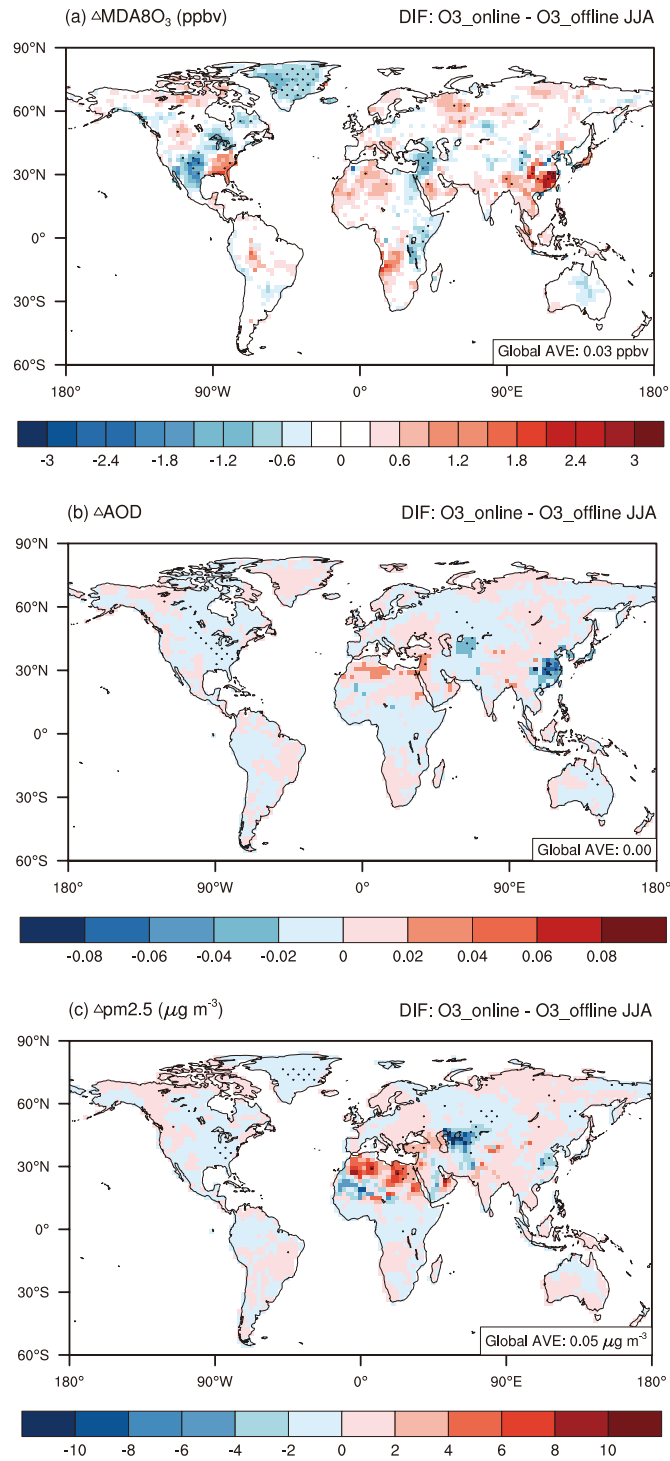
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**Figure 5.** Vertical profile of vertical velocity. Results shown are changes of the vertical velocity in (a) Eastern China and (b) Eastern US between simulations O3\_online and O3\_offline. Solid red line denotes the value 0. Please notice the differences in the scales.



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**Fig. 6.** Changes of summertime atmospheric pollution caused by O<sub>3</sub>-vegetation interactions at present day. Results shown are changes of (a) O<sub>3</sub>, (b) AOD, and (c) PM<sub>2.5</sub> between O3\_online and O3\_offline. Black dots denote areas with significant changes ( $p < 0.1$ ).

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