Global assessment of climatic responses to the ozone-vegetation interactions

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Abstract. The coupling between surface ozone (O₃) and vegetation significantly 12 13 influences regional to global climate. O3 uptake by plant stomata inhibits photosynthetic rate and stomatal conductance, impacting evapotranspiration through 14 15 land surface ecosystems. Using thea climate-vegetation-chemistry coupled model (the NASA GISS ModelE2 coupled with Yale Interactive terrestrial Biosphere, or ModelE2-16 YIBs-model,), we assess the global climatic responses to O₃-vegetation interactions 17 during boreal summer of 2010s the present day (2005-2014). High O₃ pollution reduces 18 19 stomatal conductance, resulting in-the warmer and drier conditions worldwide. The most significant responses are found in the eastern U.S. and eastern China, where local 20 latent heat flux decreases by -8.17% and -9.48%, respectively. Consequently, surface 21 22 air temperature rises increases by $+0.33\pm0.87$ °C and $+0.56\pm0.38$ °C, and respectively. 23 These temperature rises are accompanied by decreased latent heat and increased 24 sensible heat flux rises by +16.54% and +25.46% in the two hotspotboth regions. The 25 O₃-vegetation interaction also affects atmospheric pollutants. Surface maximum daily 8-hour average O_3 concentrations increase by $+1.2646\pm3.02$ ppbv in eastern China and 26 27 $+0.981.15\pm1.77$ ppbv in eastern U.S. due to the O₃-induced inhibition of stomatal uptake. With reduced atmospheric stability following the warmer climate, increased 28 cloudiness but decreased relative humidity jointly reduce aerosol optical depth 29 (AOD)by -0.06±0.01 (-14.67±12.15%) over eastern China. This study suggests that 30 31 vegetation feedback should be considered for a more accurate assessment of climatic perturbations caused by tropospheric O₃. 32

33 **1 Introduction**

34 Tropospheric ozone (O_3) , one of the most detrimental air pollutants (Myhre et al., 35 2013), not only poses threats to human health (Norval et al., 2011; Nuvolone et al., 2018) but also induces phytotoxic effects to vegetation (Mills et al., 2007; Pleijel et al., 2007). 36 When exposed to certain levels of O₃, plant photosynthesis and stomatal conductance 37 is inhibited due to the O₃ oxidation of cellular, enzyme, and chlorophyll (Dizengremel, 38 2001; Fiscus et al., 2005; Jolivet et al., 2016). Consequently, the carbon assimilation of 39 40 terrestrial ecosystems is limited (Yue and Unger, 2014; Oliver et al., 2018) and the landair exchange rates of water and heat fluxes are altered (Lombardozzi et al., 2015). 41

42 Experimental studies have shown that the excessive O₃ exposure reduced both plant photosynthesis and stomatal conductance (Ainsworth et al., 2012; Lombardozzi 43 44 et al., 2013). The reduction rates are dependent on the O₃ stomatal fluxes as well as the damaging sensitivities that vary among different vegetation types (Nussbaum and 45 Fuhrer, 2000; Karlsson et al., 2004; Pleijel et al., 2004). TraditionalSeveral exposure-46 47 based indexes likesuch as accumulated hourly O₃ concentrations over a threshold of 40 ppb (AOT40) are widely and sum of all hourly average concentrations (SUM00) are 48 used to assess O₃-induced vegetation damage (Fuhrer et al., 1997). However, such 49 statistical schemes fail; Paoletti et al., 2007). In addition, the flux-related PODy method 50 (phytotoxic O₃ dose above a threshold flux of y) is also widely applied to account 51 52 forconsider the dynamic adjustment of vegetation physiological processes.stomatal conductance (Buker et al., 2015; Sicard et al., 2016). Taking into account the variability 53 of plant sensitivities, different O₃ damage schemes were proposed to quantify the O₃ 54 impacts on land carbon assimilation from regional to global scales (Anav et al., 2011; 55 Lam et al., 2023; Lei et al., 2020). For example, Sitch et al. (2007) calculated the 56 57 simultaneous damages to both photosynthesis and stomatal conductance based on the instantaneous O3 stomatal uptake. In contrast, Lombardozzi et al. (2012) estimated the 58 inconsistent decoupled reductions in plant photosynthesis and stomatal conductance 59 using different response relationships to the cumulative O₃ stomatal uptake. 60 61 Applications of different schemes resulted in a wide range of reductions in gross primary productivity (GPP) by 2-12% globally with regional hotspots up to 20-30% 62

63 (Lombardozzi et al., 2015; Unger et al., 2020; Zhou et al., 2024).

64 The O₃-induced inhibition in stomatal conductance decreases dry deposition and consequently enhances surface O₃ concentrations (Clifton et al., 2020; Wesely and 65 Hicks, 2000; Zhang et al., 2006). Using the Sitch et al. (2007) scheme with high O₃ 66 damaging sensitivities in the elimate model ModelE2-YIBs, ModelE2-YIBs (NASA 67 GISS ModelE2 coupled with Yale Interactive terrestrial Biosphere model), Gong et al. 68 (2020) revealed that O₃-vegetation interactions increased regional O₃ concentrations by 69 70 1.8 ppbv in the eastern U.S., 1.3 ppbv in Europe, and 2.1 ppbv in eastern China for the year 2010. As a comparison, Sadiq et al. (2017) found a consistent but consistently 71 72 stronger-positive feedback on O₃ concentrations in these polluted regions using the scheme of Lombardozzi et al (2012) embedded in a different climate model. 73 74 Inclusion the Community Earth System Model (CESM). Moreover, the inclusion of 75 online O₃-vegetation interactions in numerical models will cause stronger damages toalso result in a greater loss of simulated land carbon assimilation due to the feedbacks 76 of both ecosysteme cosystems and surface O₃. This is attributable to several factors. On 77 78 one hand, the O₃ damages to leaf photosynthesis inhibit plant growth and decrease leaf area index (LAI), leading to higher reduction percentage in GPP compared to 79 simulations without LAI changes (Yue et al., 2020). On the other hand, the O₃ 80 enhancement due to vegetation feedback may cause additional vegetation damage and 81 82 result in further GPP losses (Lei et al., 2021). As a result, the O3-vegetation interactions should be considered in the global estimate of O₃ damages to ecosystem functions. 83

In addition to affecting surface O₃, the O₃-vegetation interaction can also alter the 84 water and energy exchange between land and atmosphere- through the modulation of 85 86 stomatal conductance. For example, Lombardozzi et al. (2015) used the Community 87 Land Model (CLM) and estimated that the cumulative uptake of O_3 by the leaves resulted in reduction of 2.2% in transpiration but increase of 5.4% in runoff globally. 88 Arnold et al. (2018) used the Community Earth System Model (CESM)CESM and 89 90 found that plant exposure to O3 could decrease the land-air moisture fluxes and 91 atmospheric humidity, which further reduced shortwave cloud forcing in polluted 92 regions and induced widespread surface warming up to +1.5 K. Two recent studies

utilized the WRF-chem model and revealed considerable warming and the associated
meteorological perturbations due to the O₃-vegetation interactions in China (Zhu et al.,
2022; Jin et al., 2023). However, all these modeling studies applied the same O₃
vegetation damage scheme proposed by Lombardozzi et al. (2012). It's necessary to
assess the climatic responses to O₃-vegetation interactions using different schemes so
as to explore the robust responses and the associated uncertainties.

99 In this study, we quantified the global impacts of O₃-vegetation interaction on 100 climatic conditions and surface air pollutants during 2010s using the Earth system model NASA GISS ModelE2 coupled with Yale Interactive terrestrial Biosphere 101 102 (ModelE2-YIBs) model (Yue and Unger, 2015). This fully coupled framework was implemented with the semi-mechanistic O₃ damage scheme proposed by Sitch et al. 103 104 (2007), which calculated aggregate aggregated O3 damage to photosynthesis based on varied sensitivities to instantinstantaneous stomatal O₃ uptake for across eight plant 105 functional types (PFTs). We performed sensitivity experiments to quantify the 106 107 responses of surface air temperature and precipitation to O₃-vegetation interaction. The 108 feedbacks to aerosols and O₃ concentrations were also examined.

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110 **2 Method**

111 **2.1 Model descriptions**

112 The ModelE2-YIBs is a fully coupled climate-carbon-chemistry model combining the NASA GISS ModelE2 with the YIBs vegetation model. ModelE2 is a general 113 circulation model with the horizontal resolution of $2^{\circ} \times 2.5^{\circ}$ in latitude and longitude 114 and 40 vertical layers up to 0.1 hPa. It dynamically simulates gas-phase chemistry (NOx 115 - HO_x - O_x - CO - CH4 - NMVOCs), aerosols (sulfate, nitrate, black and organic carbon, 116 dust, and sea salt), and their interactions (Menon and Rotstayn, 2006). Both the physical 117 and chemical processes are calculated every 0.5 h and the radiation module is called 118 every 2.5 h. The radiation module includes direct and indirect aerosol radiative effects 119 and accounts for absorption of multiple greenhouse gases (GHGs). For cloud optical 120 121 parameters, it uses Mie scattering, ray tracing, and matrix theory (Schmidt et al., 2006). 122 The model outperforms 20 other IPCC-class climate models in simulating surface solar radiation (Wild et al., 2013) and has been extensively validated for meteorological and
hydrological variables against observations and reanalysis data (Schmidt et al., 2014).

125 The YIBs model employs the well-established Farquhar model for leaf 126 photosynthesis and Ball-Berry model for stomatal conductance (Farquhar et al., 1980; 127 Ball et al., 1987) as follows:

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

128 <u>Here, the total leaf photosynthesis, denoted as A_{tot} (µmol m⁻² [leaf] s⁻¹), is calculated 129 considering both C₃ (Collatz et al., 1991) and C₄ plants (Collatz et al., 1992). The A_{tot} 130 is derived from the minimum value of the constraints. The ribulose-1,5-bisphosphate 131 carboxylase (Rubisco) limited rate of carboxylation is J_c :</u>

$$g_{\overline{s}} = m \frac{(A_{tot} - R_d) \times RH}{c_{\overline{s}}} + bJ_c =$$

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

132 <u>The Here, the total leaf photosynthesis, denoted as A_{tot} , is calculated as the minimum</u> 133 value among the ribulose-1,5-bisphosphate carboxylase-limited rate of carboxylation 134 (J_e) , rate restricted by the availability of light-limited rate (J_e) , and _ is J_e :

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

135 <u>The export-limited rate (J_s) . for C₃ plants and the phosphoenolpyruvate carboxylase</u>

136 (PEPC) limited rate of carboxylation for C_4 plants are represented by J_s :

$$J_{s} = \begin{cases} 0.5 \ V_{cmax} & \text{for } C_{3} \text{ plant} \\ K_{s} \times V_{cmax} \times \frac{c_{i}}{P_{atm}} & \text{for } C_{4} \text{ plant} \end{cases}$$
(4)

137 In these functions, V_{cmax} (µmol m⁻² s⁻¹) is the maximum carboxylation capacity. c_i 138 and O_i (Pa) represent the internal leaf CO₂ and oxygen partial pressure. Γ_* (Pa) 139 denotes the CO₂ compensation point, while K_c and K_o (Pa) are Michaelis–Menten 140 constants for the carboxylation and oxygenation of Rubisco, respectively. The 141 parameters Γ_* , K_c , and K_o vary with temperature based on the sensitivity of the 142 vegetation to temperature (Q₁₀ coefficient). *PAR* (µmol m⁻² s⁻¹) is the absorbed 143 photosynthetically active radiation, a_{leaf} is leaf-specific light absorbance that 144 considers sunlit and shaded leaves, and α is quantum efficiency. P_{atm} (Pa) represents 145 the ambient pressure. K_s is set to 4000 as a constant following Oleson et al. (2010), to 146 limit photosynthesis of C₄ plants get saturated at lower CO₂ concentrations.

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

The stomatal conductance (g_s) , mol [H₂O] m⁻² s⁻¹) is linked to the variations of A_{tot} 147 with parameters such as dark respiration rate $(R_d)_{\overline{r}, \mu mol m^{-2} s^{-1}}$, relative humidity 148 149 (RH), and CO₂ concentration at the leaf surface (c_s) . The model simulates the 150 biophysical processes of eight PFTs including tundra, C₃/C₄ grass, shrubland, deciduous 151 broadleaf forest, evergreen broadleaf forest, evergreen needleleaf forest, and cropland. 152 Different values are assigned to parameters m and b for each PFT (Table S1). The carbon uptake by the leaf is then accumulated and allocated to different organs to 153 154 support the plant development with dynamical changes in LAI and tree growth.

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156 **2.2 The O₃-vegetation damage scheme**

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

$$A_{totd} = A_{tot} \cdot F \tag{36}$$

$$g_{sd} = g_s \cdot F \tag{47}$$

161 where A_{totd} and g_{sd} are the unaffected photosynthesis and stomatal conductance 162 separately. The factor F is defined as:

$$F = 1 - a_h \cdot \max\left[F_{03} - F_{03,crit}, 0.0\right]$$
(58)

163 $a_h \pmod{\text{mmol m}^{-2} \text{ s}^{-1}}$ is the high O₃ sensitivity coefficient, <u>calibrated by Sitch et al. (2007)</u> 164 <u>on data from field observations by Karlsson et al. (2004)</u> and <u>Pleijel et al. (2004) to</u> 165 <u>represent 'high' sensitivity of relative species of each PFT.</u> $F_{O3,crit} \pmod{\text{m}^{-2} \text{ s}^{-1}}$ is the 166 specific threshold for O₃ damages, both of which varies with vegetation types (Table 167 S1).

$$F_{O3} = \frac{[O_3]}{R_a + [\frac{k_{O3}}{g_{sd}}]},\tag{69}$$

where $[O_3]$ represents surface O₃ concentrations, R_a (s m⁻¹) stands for the 168 169 aerodynamic and boundary layer resistance, which expresses turbulent transport efficiency in transferring sensible heat and water vapor between the land surface and a 170 171 <u>reference height</u>. The constant $k_{03}=1.67$ is the ratio of stomatal resistance for O_{3_2} 172 estimated based on the theoretical stomatal resistance to that for water. water (Laisk et 173 al., 1989). When plants are exposed to $[O_3]$ (Eq. 9), A_{tot} and g_s will decrease (Eq. 6) 174 and Eq. 7) if the excess O₃ enters leaves (Eq. 8). The increased stomatal resistance acts to protect plants by reducing the O₃ uptake of stomata. Consequently, the damage 175 scheme describes both changes in photosynthetic rate and stomatal conductance. 176

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178 **2.3 Experiments**

179 To explore the coupled O_3 -vegetation effect, we performed two sets of simulations using the ModelE2-YIBs model. The control experiment "10NO3O3 offline" was 180 181 conducted without the O₃ damages to vegetation. As a comparison, the sensitivity 182 experiment "10HO3O3 online" contained online O₃-vegetation interaction with high 183 O₃ sensitivity. For both experiments, the 2010s anthropogenic emissions of 2010 (the average of 2005-2014) for 8 species (BC, OC, CO, NH₃, NO_x, SO₂, Alkenes, and 184 185 Paraffin) from 8 economic sources (agriculture, energy, industry, transportation, resident, solvent, waste, and international shipping) and biomass burning source were 186 collected from the Coupled Model Intercomparison Project phase 6 (CMIP6) (van 187 188 Marle et al., 2017; Hoesly et al., 2018). The ensemble mean of monthly sea surface temperature (SST) and sea ice fraction (SIC) simulated by 21 CMIP6 models during 189 190 the time period 2005-2014 was employed as the boundary conditions. The cover 191 fraction of 8 PFTs (Fig. S1) fixed at 2010 were adopted from the land use harmonization 192 (LUH2) dataset (Hurtt et al., 2020). For each time-slice simulation, the model was run for 30 years with all the input data fixed and the first 10 years are used as the spin up. 193 194 We calculated the average of the last 20 years and focused on the boreal summer season 195 (June-July-August, JJA) when the interaction of vegetation and surface O₃ reaches the maximum in one year- (fig. S3). In order to show the uncertainty introduced by the internal variability of the model, all the related global/regional values are denoted as "mean/sum \pm standard deviation of the last 20 model years". We explored the climatic responses to O₃-vegetation interactions as the differences between "10HO3O3_online" and "10NO3O3_offline" on the global scale with the special focus over the hotspot regions such as eastern U.S. (30–40° N, 80–90 ° W) and eastern China (22.5–38° N, 106–122° E).

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204 **2.4 Data for** evaluations model evaluation

We evaluated the simulated air pollutants, carbon fluxes, and meteorological 205 variables from 'O3 offline' run using observational and reanalysis datasets. The 206 207 worldwide observations of Θ_3 the maximum daily 8-hour average O_3 (MDA8 O_3) concentrations were <u>mainly</u> collected from three regional networks: Air Quality 208 Monitoring Network operated by Ministry of Ecology and Environment (AQMN-MEE) 209 in China, the Clean Air Status and Trends Network (CASTNET) in the U.S., and the 210 211 European Monitoring and Evaluation Programme (EMEP) in Europe. For the latter two networks, we chose the average over 2009-2011, while for Observations used for 212 213 validation beyond China, sourced from Sofen et al. (2016), are averaged over the period 2005-2014. This dataset encompasses 7288 station records worldwide and excludes the 214 215 uncertainty associated with high mountain-top sites. For AQMN-MEE, the mean value of 2014-2018 was used due to its establishment in 2013. The simulated aerosol optical 216 217 depth (AOD) was and LAI were validated using satellite-based data from the Moderate Resolution Imaging Spectroradiometer (MODIS) retrievals collection 5 (Remer et al., 218 2005) (http://modis.gsfc.nasa.gov/) averaged for the years 2009-20112005-2014. The 219 220 simulated GPP was evaluated against the data product upscaled from the FLUXNET eddy covariance measurements for 2009-2011 (Jung et al., 20092011). The daily 221 temperature at 2m (T_{2m}) in $\frac{2009-2011}{2005-2014}$ was obtained from the National 222 Centers for Environmental Prediction/National Center for Atmospheric Research 223 224 (NCEP/NCAR) reanalysis 1 (NCEP1) (Kalnay et al., 1996). For precipitation, we used the monthly data averaged in 2005-2014 from Global Precipitation Climatology Project 225

(GPCP) (Huffman et al., 1997; Adler et al., 2018). All these datasets were interpolated
 to the same resolution as ModelE2-YIBs model. NormalizedRoot-mean-square-error
 (RMSE) and normalized mean biases (NMBs) were applied to quantify the deviations
 of simulations from observations-as follows:

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (S_i - O_i)^2}$$
(10)
$$NMB = \frac{\sum_{i=1}^{n} (S_i - O_i) / \sum_{i=1}^{n} O_i}{100\% NMB} = \sum_{i=1}^{n} (S_i - O_i) / \sum_{i=1}^{n} O_i \times$$

100%

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

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234 **3. Results**

235 **3.1 Model** The control simulation and model evaluations

236 We first evaluated the air pollutants simulated by the control simulation O3 offline of ModelE2-YIBs model (Fig. 1). Over a total of 491503 grids with site-level O3 237 238 measurements (Fig. 1b), the model adequately replicated both the magnitude and spatial 239 distribution of the maximum daily 8-hour average (MDA8) O₃-concentrations ([_O₃])₂ with correlation coefficient (r) of 0.5859 and NMB of -1.272.54% (Fig. 1c). Simulated 240 summertime surface [MDA8 O₃] was high in regions with large anthropogenic 241 242 emissions, such as western Europe and eastern China (Ohara et al., 2007), as well as in central Africa with frequent fire emissions (van der Werf et al., 2017). On the global 243 scale, the model yielded an average [MDA8 O₃] of 44.3643.93 ppbv and observations 244 245 showed an average of 44.5772 ppbv over the same grids. However, the modeled result is overestimated over the North China Plain and slightly underestimated over the U.S-... 246 247 likely due to the biases in the emission inventories and predicted climate that drive the O₃ production. Simulated AOD at 550 nm by O₃ offline (Fig. 1d) showed similar 248 spatial pattern as the satellite retrievals (Fig. 1e) with a high R=0.7775 and low NMB 249 of -6.277.35% globally (Fig. 1f). Both the simulations and observations showed AOD 250 251 hotspots over North Africa and the Middle East where dust emissions dominate, and in northern India and eastern China where anthropogenic emissions are large<u>- (Feng et al.,</u>
253 <u>2020).</u>

254 We then evaluated the simulated GPP and LAI by the control experiment for the boreal summer period (Fig. 2). Observations showed GPP hotspots over boreal forests 255 such as eastern U.S., Eurasia, and East Asia and the tropical forests such as Amazon, 256 central Africa, and Indonesia (Fig. 2b). The seasonal total GPP was estimated to be 257 41.63Pg[C], which accounted for 35% of the annual amount. Simulations well-captured 258 259 the observed GPP pattern on the global scale, with r = 0.6364 and NMB = -12.447.81%over 2581 grids (Fig. 2c), with underestimation in the tundra area and slight 260 overestimation in the tropical rain forest and evergreen forest regions. The model 261 simulated a seasonal total GPP of 36.4538.69 Pg[C], equivalent to 34% of the annual 262 263 amount. Simulated LAI showed similar patterns as GPP (Fig. 2d) and resembled observed LAI (Fig. 2e) with a high-spatial correlation r = 0.79 and a low NMB = -264 5.1943% over 4435 grids globally (Fig. 2f). 265

We further validated the simulated meteorology from O3 offline (Fig. S2). For 266 267 surface air temperature, the model (Fig. S2a) reproduced observed (Fig. S2b) pattern with low NMBRMSE of 8.49%3.21 °C and high r of 0.99 against observations (Fig. 268 S2c). For precipitation, both simulations the simulation (Fig. S2d) and 269 observationscaptures the observed spatial pattern (Fig. S2e) showed high values in the 270 tropical oceans with NMB = $\frac{16.9117.26}{17.26}$ % and r = 0.74 between them 75 (Fig. S2f). 271 Overall, the model showed good performance in the simulations captures the spatial 272 characteristics and magnitudes of air pollutants, biospheric parameters, and 273 meteorological fields, and provided making it a useful valuable tool for studying the-O3-274 275 vegetation interactions.

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3.2 O₃ damage to terrestrial ecosystems

We assessed the damaging effects of surface O_3 to ecosystems (Fig.due to online O_3 -vegetation interactions (Fig. 3). The impacts of O_3 on biospheric variables were mainly located in regions characterized by abundant vegetation cover and elevated O_3 concentrations. On the global scale, O_3 induced the GPP reduction of -<u>1.80±0.8761</u> PgC 282 yr⁻¹ (-3.094.69±1.56%, Fig. 3a). This deleterious effect was more pronounced in 283 specific regions, notably eastern China and eastern U.S., with significant GPP declines of $-\frac{18.43}{25.40\pm1.90}$ % and $-\frac{16.12}{20.14\pm5.02}$ %, respectively, under high O₃ sensitivity 284 conditions (Fig. 3a and Table S2). Meanwhile, stomatal conductance significantly 285 decreased in the middle latitudes of Northern Hemisphere (Fig. 3b). The most 286 substantial relative change of -30.62 ± 4.30 % was observed in eastern China, followed 287 by -25.65±9.32% in the eastern U.S. (Fig. 3b and Table S2). Though there are positive 288 289 responses in some regions, they are not dominant and hardly significant. These values were stronger than that for GPP (Fig. 3a), likely due to the climatic feedback to O₃-290 vegetation interactions. The opening of plant stoma plays a crucial role in regulating 291 the energy and water exchange between land surface and the atmosphere. The inhibition 292 293 of stomatal conductance by surface O₃ leads to the warmer (Fig. 4a) and drier (Fig. 4b) climate in those hotspot regions, resulting in even stronger inhibition effects on stomatal 294 conductance. Following the changes in GPP, global LAI on average decreased by 295 $0.01\pm0.01 \text{ m}^2 \text{ m}^{-2}$ (-0.62±0.84%) with regional maximums of -4.53±1.14% in eastern 296 China and $-5.87\pm3.11\%$ in eastern U.S. (Table S2). 297

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3.3 Global climatic responses to O₃-vegetation interactions

In response to the O₃-induced inhibition of stomatal conductance, surface air 300 temperature increased by $0.05 \times C_{05\pm 0.20}$ (Fig. 4a) while precipitation decreased by 301 -0.01 ± 0.03 mm day⁻¹ (Fig. 4b) on the global scale. The most significant change was the 302 warming of 0.56 ± 0.38 °C and precipitation reduction of -0.79 ± 1.05 mm day⁻¹ (-303 304 16.18±20.38%) in eastern China (Table S3), following the largest inhibition to stomatal 305 conductance (Fig. 3b). Such warming and rainfall deficit also appeared in eastern U.S. 306 and western Europe, where the O_3 -vegetation interactions were notable. The O_3 -307 induced inhibition to stomatal conductance decreased latent heat flux (Fig. 4e) and the 308 consequent precipitation (Fig. 4b) in those hotspot regions. Meanwhile, the reduction 309 of latent heat flux promotes surface air temperature (Fig. 4a), resulting in the increase 310 of sensible heat flux (Fig. 4f). Such warming was also reported in field experiments,

where relatively high O₃ exposure resulted in noticeable increases of canopy temperature along with reductions of transpiration (Bernacchi et al., 2011; VanLoocke et al., 2012). Globally, temperature and precipitation showed patchy responses with both positive and negative anomalies, suggesting that the regional hotspots of O₃induced meteorological changes propagate to surrounding areas through atmospheric perturbations.

We further examined the changes in air humidity and cloudiness. Surface relative 317 humidity decreased by $-0.18\pm0.53\%$ globally with a similar pattern as that of 318 precipitation (Fig. 4c). The most significant reductions were over eastern China and 319 320 eastern U.S., where both the warming (Fig. 4a) and rainfall deficit (Fig. 4b) contributed to the drought. However, in the adjacent regions such as northern China and central 321 322 U.S., both rainfall and surface relative humidity showed certain enhancement. These changes were associated with the regional increase of cloud cover (Fig. 4d). The 323 sensible heat flux increased by 6.3 ± 5.4 W m⁻² (16.54±15.59%) and 7.12 ± 3.86 W m⁻² 324 (25.46±14.71%) in eastern U.S. and eastern China, respectively, suggesting a transfer 325 326 of thermal energy from land to the atmosphere by O₃-vegetation interactions (Fig. 4f and Table S3). The warming effect further triggered anomalous updrafts in the lower 327 328 troposphere, represented by the changes in vertical velocity (Fig. 5), leading to enhanced convection, reduced atmospheric stability, and consequently an increase in 329 low-level cloudiness (Fig. 4d). However, despite the usual cooling effect associated 330 with increased cloud cover due to reductions in radiation, in regions predominantly 331 332 influenced by O_3 -vegetation interactions, this cooling effect was outweighed by the O_3 induced warming through inhibition of stomatal conductance. Therefore, temperatures 333 exhibited an overall increase of 0.56 ± 0.38 °C in eastern China and 0.33 ± 0.87 °C in the 334 335 eastern U.S. (Table S3).

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337 3.4 Changes of air pollution by O₃-vegetation interactions

Changes in surface water and heat fluxes induced by O_3 -vegetation interactions could feed back to affect air pollutants such as O_3 and aerosols. As Fig. 6a and Table S4 show, surface MDA8_O₃ concentrations enhanced 1.<u>2646±3.02</u> ppbv in eastern 341 China and $0.981.15\pm1.77$ ppbv in eastern U.S. due to the decreased dry deposition 342 following O₃ inhibition on stomatal conductance. It indicates that the high 343 contemporary O₃ pollution may worsen air quality through O₃-vegetation interactions. However, negative O₃ changes were predicted in central U.S. and western China, where 344 the increased rainfall dampened O₃ through chemical reactions and wet deposition. On 345 a global scale, surface $\underline{MDA8}$ O₃ showed a limited increase of 0.0203 ± 0.4 ppbv due to 346 the offset between positive and negative feedbacks. The enhancement of O₃ 347 348 concentrations in polluted regions may exacerbate the warming effect of O₃ as a 349 greenhouse gas and cause additional damages to vegetation. For instance, offline O₃ damages on GPP in eastern China and the eastern US are -0.52±0.03 Pg[C] (-350 24.98±0.91%) and -0.17±0.02 Pg[C] (-16.71±1.16%), respectively, smaller than those 351 352 induced by O₃-vegetation interactions (Table S2).

353 Aerosols also exhibited evident changes by the O₃-vegetation interactions. The AOD showed significant reductions over the hotspot regions such as eastern China and 354 eastern U.S. (Fig. 6b). In the ModelE2-YIBs model, sulfate was especially sensitive to 355 356 cloud which could enhance the aerosol scavenging through cloud water precipitation (Koch et al., 2006). The large enhancement of cloudiness removed sulfate more 357 358 efficiently than other aerosol species, leading to an average decline of -1.94 ± 1.67 µg m^{-3} (-8.52±6.88%) in PM_{2.5} loading over eastern China (Fig. S3S4 and Table S4). 359 Meanwhile, the reduction of surface relative humidity (Fig. 4c) in the regions with 360 strong O₃-vegetation interactions limited the hygroscopic growth of aerosols, leading 361 362 to a more noticeable decrease in AOD (Petters and Kreidenweis, 2007; Revised algorithm for estimating light extinction from IMPROVE particle speciation data, 363 <u>2023Pitchford et al., 2007</u>) by -0.06 \pm 0.05 (-14.67 \pm 16.75%) in eastern China (Table S4). 364 The similar aerosol changes were found in eastern U.S. but with smaller reductions of 365 $PM_{2.5}$ by $-0.27 \pm 0.36 \ \mu g \ m^{-3}$ (-6.01 $\pm 7.9\%$) and AOD by $-0.01 \pm 0.01 \ (-8.2515 \pm 9.38\%)$ 366 (Table S4). Beyond the key O₃-vegetation coupling regions, positive but insignificant 367 changes in AOD were predicted, leading to the moderate AOD changes on the global 368 369 scale (Fig. 6b).

371 4. ConclusionsDiscussion and discussionconclusions

372 We examined the O₃-vegetation feedback to climate and air pollution in the 2010s 373 using the fully coupled climate-carbon-chemistry model ModelE2-YIBs. During boreal 374 summer, surface O₃ resulted in strong damages to GPP and inhibitions to stomatal conductance with regional hotspots over eastern China and eastern U.S. Consequently, 375 376 surface transpiration was weakened, leading to decreased latent heat fluxes and relative 377 humidity but increased surface air temperature. Meanwhile, the surface warming 378 increased cloud cover by reducing atmospheric stability. The However, the 379 enhancement of cloudiness further decreased surface temperature and promoted precipitation nearby outside the key regions with intense O₃-vegetation interactions. The 380 O₃-induced inhibition to stomatal conductance resulted in a localized increase in O₃ 381 382 concentrations. In contrast, the increased cloud cover and decreased relative humidity jointly reduced AOD in hotspot regions. On the global scale, the mean changes of both 383 climate and air pollution were moderate due to the offset between the changes with 384 opposite signs. 385

386 Our predicted changes in water/heat fluxes by O₃-vegetation interactions were consistent with previous studies (Lombardozzi et al., 2015; Arnold et al., 2018; Gong 387 388 et al., 2020). For example, the simulations by Lombardozzi et al. (2015) revealed that 389 surface O_3 reduces global GPP by 8%-12% and transpiration by 2-2.4% with regional 390 reductions up to 20% for GPP and 15% for transpiration in eastern China and U.S. These changes were in general consistent with our results though we predicted larger 391 392 reductions in transpiration than GPP due to O₃-vegetation interactions. Using the same scheme as Lombardozzi et al. (2015), Sadiq et al. (2017) showed that O₃-vegetation 393 coupling induced the surface warming of 0.5-1°C and O3 enhancement of 4-6 ppbv in 394 395 eastern China and eastern U.S. The magnitude of these responses was much stronger 396 than our predictions, likely because they considered the accumulation effect of O₃. In 397 contrast, the regional simulations by Jin et al. (2023) revealed that O₃-vegetation 398 coupling led to the increases of temperature up to 0.16°C and surface O₃ up to 0.6 ppbv 399 in eastern China, both of which were smaller than our predictions. The damage scheme they use, which depends on cumulative O_3 uptake, omits the difference in impact on sunlit or shaded leaves and will overestimate the O_3 damage on GPP compared to the scheme we use, which considers transient O_3 flux (Cao et al., 20232024). The discrepancies of O_3 -vegetation feedback using the same O_3 damage schemes revealed the uncertainties from climate and chemistry models. Our predictions were within the range of previous estimates for both climatic and O_3 changes.

There were some limitations in our simulated O₃-vegetation interactions. First, the 406 407 semi-mechanistic O3 damage scheme we used in the study linked the damages to 408 photosynthesis with those to stomatal conductance (Sitch et al., 2007), leading to 409 stronger inhibition percentage in stomatal conductance than that in photosynthesis considering the O₃-vegetation feedback. However, some observations showed that the 410 411 damage to stomatal conductance occurred more slowly and might not be proportional to the decline of photosynthetic rates (Gregg et al., 2006; Lombardozzi et al., 2012). 412 Second, observations have shown large variability of plant sensitivities to O₃ damages. 413 414 The Sitch et al. (2007) scheme employed the low to high ranges of sensitivity to indicate 415 the inter-specific variabilities. In this study, we employed only the high O₃ sensitivity to explore the maximum responses. The possible uncertainties due to varied O₃ damage 416 417 sensitivities deserved further investigations. Third, large-scale observations were not 418 available to validate the simulated regional to global responses of climate and air 419 pollutants. The O₃ vegetation damage scheme was extensively validated against site-420 level measurements of both photosynthesis (Yue and Unger, 2018) and stomatal 421 conductance (Yue et al., 2016). However, we were conservative about the derived 422 global responses given that previous studies showed large discrepancies using the same 423 O₃ damage scheme but implemented in different climate and/or chemistry models (Lombardozzi et al., 2015; Sadiq et al., 2017; Jin et al., 2023). Furthermore, the 2°×2.5° 424 resolution of current ModelE2-YIBs has limitation due to the high computational 425 demands. Ito et al. (2020) shows that the ModelE2.1 with fixed vegetation traits 426 reproduces carbon fluxes well, and that the model results are involved in the CMIP6 427 428 Coupled Climate-Carbon Cycle MIP (C4MIP). However, analysis of the climate model shows that high-resolution exhibits models exhibit improved simulations of extreme 429

430 events (Chang et al., 2020; Ban et al., 2021), and the application of which have certain 431 effect on O₃-vegetation interactions (Mills et al., 2016; Lin et al., 2020). While chemical 432 transport model shows that models with relatively coarse resolution can raise biases in 433 simulated air pollutants, though it captures thethey still capture large-scale general pattern almost the same aspatterns similar to fine-resolution results and is reasonable as 434 compared to observational data (Wang et al., 2013; Li et al., 2016; Lei et al., 2020). 435 Moreover, we omit the slow climatic feedback caused by air-sea interaction in the 436 437 simulations. Studies have revealed that these interactions may result in different 438 climatic perturbations from those simulations with fast responses of land surface alone (Yue et al., 2011). A dynamic ocean model is considered to enrich the future research. 439 440 Meanwhile, this study does not isolate the different impacts of aerosols, even though 441 the radiation module includes both direct and indirect radiative effects. We will 442 investigate this further in the future by identifying the main processes.

443 Despite these uncertainties, our simulations revealed considerable changes of both 444 climate and air pollutants in response to O₃-vegetation interactions. The most intense 445 warming, dryness, and O₃ enhancement were predicted in eastern China and eastern U.S., affecting the regional climate and threatening public health for these top two 446 447 economic centers. In contrast, we for the first time revealed the reduction of aerosol loading in those hotspot regions, suggesting both positive and negative effects to air 448 449 pollutants by O₃-vegetation feedback. Such interactions should be considered in the 450 Earth system models so as to better project future changes in climate and air pollutants following the anthropogenic interventions to both O₃ precursor emissions and 451 452 ecosystem functions.

454	Data Availability
455	The observational data and model outputs that support the findings in this study are
456	available from corresponding authors upon reasonable request.
457	
458	Author contributions
459	XY conceived the project. XZ performed the model simulations, conducted results
460	analysis and wrote the draft manuscript. XY, CT and XL assisted in the interpretation
461	of the results and contributed to the discussion and improvement of the paper.
462	
463	Competing interests
464	The authors declare that they have no conflict of interest.
465	
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Figure 1. Evaluation of the boreal summertime (June-August) air pollutants in 2010sat 473 the present day simulated by the ModelE2-YIBs model. Surface daily maximum 8-hour 474 ozone (MDA8 O₃, upper; a-c) and aerosol optical depth (AOD, bottom; d-f) from the 475 simulation 10NO3 (leftO3 offline (a & d) and observations (middleb & e) are 476 compared. The correlation coefficients (r), root mean square error (RMSE), normalized 477 478 mean bias (NMB), and number of grid cells (n) for the comparisons are listed on the 479 scatter plots (e & f). The dashed line denotes the 1 : 1 ratio. The red line is the linear regression between the simulation and observation.mean bias maps (c & f). 480



486 <u>c</u>) and leaf area index (LAI, bottom panels; <u>d-f</u>).







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Figure 3. Changes of boreal summertime biospheric variables induced by O_3 -damages in 2010s.-vegetation interactions at the present day. Results shown are changes of (a) GPP, (b) canopy conductance, and (c) LAI between simulations 10HO3O3_online and 10NO3O3_offline. Black dots denote areas with significant changes (p < 0.1). Please notice the differences in the color scales.





Figure 4. Changes of boreal summertime meteorological fields by $ozoneO_3$ -vegetation interactions in 2010sat the present day. Results shown are changes of (a) surface_air temperature, (b) precipitation, (c) surface relative humidity, (d) low level cloudiness, (e) latent heat flux, and (f) sensible heat flux between simulations 10HO3O3_online and 10NO3O3_offline. For heat fluxes, positive values (shaded in red color) indicate the upward fluxes change. Black dots denote areas with significant changes (p < 0.1). Please notice the differences in the color scales.







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Fig. 6. Changes of summertime atmospheric pollution caused by $ozoneO_3$ -vegetation interactions in 2010s.at present day. Results shown are changes of (a) $ozoneO_3$, (b) AOD, and (c) PM_{2.5} between 10HO3O3_online and 10NO3O3_offline. Black dots denote areas with significant changes (p < 0.1). Please notice the differences in the color scales.

518 **References**

- Adler, R. F., Sapiano, M. R. P., Huffman, G. J., Wang, J.-J., Gu, G., Bolvin, D., Chiu,
 L., Schneider, U., Becker, A., Nelkin, E., Xie, P., Ferraro, R., and Shin, D.-B.: The
 Global Precipitation Climatology Project (GPCP) Monthly Analysis (New Version
 and a Review of 2017 Global Precipitation, Atmosphere, 9, 138,
 https://doi.org/10.3390/atmos9040138, 2018.
- Ainsworth, E. A., Yendrek, C. R., Sitch, S., Collins, W. J., and Emberson, L. D.: The
 effects of tropospheric ozone on net primary productivity and implications for
 climate change, Annu. Rev. Plant. Biol., 63, 637–661,
 https://doi.org/10.1146/annurev-arplant-042110-103829, 2012.
- Anav, A., Menut, L., Khvorostyanov, D., and Viovy, N.: Impact of tropospheric ozone
 on the Euro-Mediterranean vegetation, Glob. Change. Biol., 17, 2342–2359,
 https://doi.org/10.1111/j.1365-2486.2010.02387.x, 2011.
- 531 Arnold, S. R., Lombardozzi, D., Lamarque, J. -F., Richardson, T., Emmons, L. K., Tilmes, S., Sitch, S. A., Folberth, G., Hollaway, M. J., and Val Martin, M.: 532 533 Simulated Global Climate Response to Tropospheric Ozone-Induced Changes in 13070-13079. 534 Plant Transpiration, Geophys. Res. Lett., 45, https://doi.org/10.1029/2018GL079938, 2018. 535
- Ball, J. T., Woodrow, I. E., and Berry, J. A.: A Model Predicting Stomatal Conductance
 and its Contribution to the Control of Photosynthesis under Different
 Environmental Conditions, in: Progress in Photosynthesis Research, edited by:
 Biggins, J., Springer Netherlands, Dordrecht, 221–224,
 https://doi.org/10.1007/978-94-017-0519-6_48, 1987.
- Ban, N., Caillaud, C., Coppola, E., Pichelli, E., Sobolowski, S., Adinolfi, M., Ahrens,
 B., Alias, A., Anders, I., Bastin, S. and Belušić, D.: The first multi-model ensemble
 of regional climate simulations at kilometer-scale resolution, part I: evaluation of
 precipitation, Clim. Dynam., 57, 275-302, https://doi.org/10.1007/s00382-02105708-w, 2021.
- Buker, P., Feng, Z., Uddling, J., Briolat, A., Alonso, R., Braun, S., Elvira, S., Gerosa,
 G., Karlsson, P. E., Le Thiec, D., Marzuoli, R., Mills, G., Oksanen, E., Wieser, G.,
 Wilkinson, M., and Emberson, L. D.: New flux based dose-response relationships
 for ozone for European forest tree species, Environ. Pollut., 206, 163–174,
 https://doi.org/10.1016/j.envpol.2015.06.033, 2015.
- Bernacchi, C. J., Leakey, A. D. B., Kimball, B. A., and Ort, D. R.: Growth of soybean
 at future tropospheric ozone concentrations decreases canopy evapotranspiration
 and soil water depletion, Environ. Pollut., 159, 1464–1472,
 https://doi.org/10.1016/j.envpol.2011.03.011, 2011.
- 555 Cao, J., Yue, X. and Ma, M.: Simulation of ozone-vegetation coupling and feedback in

- 556 China using multiple ozone damage schemes, Atmos. Chem. Phys., 24(7), 3973557 3987, https://doi.org/10.5194/acp-24-3973-2024, 2024.
- 558 Chang, P., Zhang, S., Danabasoglu, G., Yeager, S.G., Fu, H., Wang, H., Castruccio, F.S., 559 Chen, Y., Edwards, J., Fu, D. and Jia, Y.: An unprecedented set of high-resolution earth system simulations for understanding multiscale interactions in climate 560 variability and change. Model. Earth. 561 J. Adv. Sv., 12, https://doi.org/10.1029/2020MS002298, 2020. 562
- Clifton, O. E., Paulot, F., Fiore, A. M., Horowitz, L. W., Correa, G., Baublitz, C. B.,
 Fares, S., Goded, I., Goldstein, A. H., Gruening, C., Hogg, A. J., Loubet, B.,
 Mammarella, I., Munger, J. W., Neil, L., Stella, P., Uddling, J., Vesala, T., and
 Weng, E.: Influence of Dynamic Ozone Dry Deposition on Ozone Pollution, J.
 Geophys. Res-Atmos, 125, e2020JD032398,
 https://doi.org/10.1029/2020JD032398, 2020.
- Collatz, G. J., Ball, J. T., Grivet, C., and Berry, J. A.: Physiological and Environmental Regulation of Stomatal Conductance, Photosynthesis and Transpiration a Model
 That Includes a Laminar Boundary-Layer, Agr. Forest Meteorol., 54, 107–136,
 doi:10.1016/0168-1923(91)90002-8, 1991.
- 573 <u>Collatz, G. J., Ribas-Carbo, M., and Berry, J. A.: Coupled Photosynthesis-Stomatal</u>
 574 <u>Conductance Model for Leaves of C4 Plants, Aust. J. Plant Physiol., 19, 519–538,</u>
 575 <u>https://doi.org/10.1071/PP9920519, 1992.</u>
- Dizengremel, P.: Effects of ozone on the carbon metabolism of forest trees, Plant.
 Physiol. Bioch., 39, 729–742, https://doi.org/10.1016/S0981-9428(01)01291-8,
 2001.
- Farquhar, G. D., von Caemmerer, S., and Berry, J. A.: A biochemical model of
 photosynthetic CO₂ assimilation in leaves of C₃ species, Planta, 149, 78–90,
 https://doi.org/10.1007/BF00386231, 1980.
- Feng, L., Smith, S. J., Braun, C., Crippa, M., Gidden, M. J., Hoesly, R., Klimont, Z.,
 van Marle, M., van den Berg, M., and van der Werf, G. R.: The generation of
 gridded emissions data for CMIP6, Geosci. Model Dev., 13, 461–482,
 https://doi.org/10.5194/gmd-13-461-2020, 2020.
- Fiscus, E. L., Booker, F. L., and Burkey, K. O.: Crop responses to ozone: uptake, modes
 of action, carbon assimilation and partitioning, Plant. Cell. Environ., 28, 997–1011,
 https://doi.org/10.1111/j.1365-3040.2005.01349.x, 2005.
- Fuhrer, J., Skärby, L. and Ashmore, M.R.: Critical levels for ozone effects on vegetation
 in Europe, Environ. Pollut., 97, 91-106, https://doi.org/10.1016/S02697491(97)00067-5, 1997.
- 592 Gong, C., Lei, Y., Ma, Y., Yue, X., and Liao, H.: Ozone–vegetation feedback through

- dry deposition and isoprene emissions in a global chemistry–carbon–climate model, Atmos. Chem. Phys., 20, 3841–3857, https://doi.org/10.5194/acp-20-3841-2020, 2020.
- Gregg, J. W., Jones, C. G., and Dawson, T. E.: Physiological and Developmental Effects
 of O₃ on Cottonwood Growth in Urban and Rural Sites, Ecol. Appl., 16, 2368–
 2381, https://doi.org/10.1890/1051-0761(2006)016[2368:PADEOO]2.0.CO;2,
 2006.
- Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T.,
 Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L.,
 Kholod, N., Kurokawa, J., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P.
 R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive
 gases and aerosols from the Community Emissions Data System (CEDS), Geosci.
 Model. Dev., 11, 369–408, https://doi.org/10.5194/gmd-11-369-2018, 2018.
- Huffman, G. J., Adler, R. F., Arkin, P., Chang, A., Ferraro, R., Gruber, A., Janowiak, J.,
 McNab, A., Rudolf, B., and Schneider, U.: The Global Precipitation Climatology
 Project (GPCP) Combined Precipitation Dataset, B. Am. Meteorol. Soc., 78, 5–20,
 https://doi.org/10.1175/1520-0477(1997)078<0005:TGPCPG>2.0.CO;2, 1997.
- 610 Hurtt, G. C., Chini, L., Sahajpal, R., Frolking, S., Bodirsky, B. L., Calvin, K., Doelman, 611 J. C., Fisk, J., Fujimori, S., Klein Goldewijk, K., Hasegawa, T., Havlik, P., Heinimann, A., Humpenöder, F., Jungclaus, J., Kaplan, J. O., Kennedy, J., Krisztin, 612 T., Lawrence, D., Lawrence, P., Ma, L., Mertz, O., Pongratz, J., Popp, A., Poulter, 613 B., Riahi, K., Shevliakova, E., Stehfest, E., Thornton, P., Tubiello, F. N., van 614 Vuuren, D. P., and Zhang, X.: Harmonization of global land use change and 615 management for the period 850-2100 (LUH2) for CMIP6, Geosci. Model. Dev., 616 13, 5425-5464, https://doi.org/10.5194/gmd-13-5425-2020, 2020. 617
- Ito, G., Romanou, A., Kiang, N.Y., Faluvegi, G., Aleinov, I., Ruedy, R., Russell, G.,
 Lerner, P., Kelley, M. and Lo, K.: Global carbon cycle and climate feedbacks in
 the NASA GISS ModelE2, J. Adv. Model. Earth. Sy., 12, https://doi.org/
 10.1029/2019MS002030, 2020.
- Jin, Z., Yan, D., Zhang, Z., Li, M., Wang, T., Huang, X., Xie, M., Li, S., and Zhuang,
 B.: Effects of Elevated Ozone Exposure on Regional Meteorology and Air Quality
 in China Through Ozone-Vegetation Coupling, J. Geophys. Res-Atmos., 128,
 e2022JD038119, https://doi.org/10.1029/2022JD038119, 2023.
- Jolivet, Y., Bagard, M., Cabané, M., Vaultier, M.-N., Gandin, A., Afif, D., Dizengremel,
 P., and Le Thiec, D.: Deciphering the ozone-induced changes in cellular processes:
 a prerequisite for ozone risk assessment at the tree and forest levels, Ann. For. Sci.,
 73, 923–943, https://doi.org/10.1007/s13595-016-0580-3, 2016.
- 630 Jung, M., Reichstein, M., and Bondeau, A.: Towards global empirical upscaling of

631	FLUXNET eddy covariance observations: validation of a model tree ensemble
632	approach using a biosphere model, Biogeosciences, 6, 2001–2013, 2009Margolis,
633	H.A., Cescatti, A., Richardson, A.D., Arain, M.A., Arneth, A., Bernhofer, C.,
634	Bonal, D., Chen, J. and Gianelle, D.: Global patterns of land-atmosphere fluxes of
635	carbon dioxide, latent heat, and sensible heat derived from eddy covariance,
636	satellite, and meteorological observations. J. Geophys. Res-Biogeosci., 116(G3),
637	https://doi.org/10.1029/2010JG001566, 2011.
638	Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M.,
539	Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W.,
640	Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R.,
541	Jenne, R., and Joseph, D.: The NCEP/NCAR 40-Year Reanalysis Project, B. Am.
542	Meteorol. Soc., 77, 437–472, https://doi.org/10.1175/1520-
343	0477(1996)077<0437:TNYRP>2.0.CO;2, 1996.
544	Karlsson, P., Uddling, J., Braun, S., Broadmeadow, M., Elvira, S., Gimeno, B., Le Thiec,
345	D., Oksanen, E., Vandermeiren, K., Wilkinson, M., and Emberson, L.: New critical
546	levels for ozone effects on young trees based on AOT40 and simulated cumulative
647	leaf uptake of ozone, Atmos. Environ., 38, 2283–2294,
348	https://doi.org/10.1016/j.atmosenv.2004.01.027, 2004.
649	Koch, D., Schmidt, G. A., and Field, C. V.: Sulfur, sea salt, and radionuclide aerosols
650	in GISS ModelE, J. Geophys. Res-Atmos, 111,
651	https://doi.org/10.1029/2004JD005550, 2006.
652	Laisk, A., Kull, O., & Moldau, H.: Ozone concentration in leaf intercellular air spaces
653	is close to zero. Plant Physiol., 90(3), 1163-1167,
654	https://doi.org/10.1104/pp.90.3.1163, 1989.
655	Lam, J. C. Y., Tai, A. P. K., Ducker, J. A., and Holmes, C. D.: Development of an
656	ecophysiology module in the GEOS-Chem chemical transport model version
657	12.2.0 to represent biosphere-atmosphere fluxes relevant for ozone air quality,
658	Geosci. Model. Dev., 16, 2323–2342, https://doi.org/10.5194/gmd-16-2323-2023,
659	2023.
60	Lei, Y., Yue, X., Liao, H., Gong, C., and Zhang, L.: Implementation of Yale Interactive
61	terrestrial Biosphere model v1.0 into GEOS-Chem v12.0.0: a tool for biosphere-
362	chemistry interactions, Geosci. Model. Dev., 13, 1137-1153,
663	https://doi.org/10.5194/gmd-13-1137-2020, 2020.
664	Lei, Y., Yue, X., Liao, H., Zhang, L., Yang, Y., Zhou, H., Tian, C., Gong, C., Ma, Y., and
665	Gao, L.: Indirect contributions of global fires to surface ozone through ozone-
666	vegetation feedback, Atmos. Chem. Phys., 21, 11531-11543,
667	https://doi.org/10.5194/acp-21-11531-20212021.
668	Li, Y., Henze, D.K., Jack, D.: The influence of air quality model resolution on health

669 670	impact assessment for fine particulate matter and its components, Air. Qual. Atmos. Hlth., 9, 51-68, https://doi.org/10.1007/s11869-015-0321-z, 2016.
671	Lin, M., Horowitz, L.W., Xie, Y., Paulot, F., Malyshev, S., Shevliakova, E., Finco, A.,
672	Gerosa, G., Kubistin, D. and Pilegaard, K.: Vegetation feedbacks during drought
673	exacerbate ozone air pollution extremes in Europe, Nat. Clim. Change., 10,444-
674	451, https://doi.org/10.1038/s41558-020-0743-y, 2020.
675 676 677 678	Lombardozzi, D., Levis, S., Bonan, G., and Sparks, J. P.: Predicting photosynthesis and transpiration responses to ozone: decoupling modeled photosynthesis and stomatal conductance, Biogeosciences, 9, 3113–3130, https://doi.org/10.5194/bg-9-3113-2012, 2012.
679 680 681 682	Lombardozzi, D., Sparks, J. P., and Bonan, G.: Integrating O ₃ influences on terrestrial processes: photosynthetic and stomatal response data available for regional and global modeling, Biogeosciences, 10, 6815–6831, https://doi.org/10.5194/bg-10-6815-2013, 2013.
683	Lombardozzi, D., Levis, S., Bonan, G., Hess, P. G., and Sparks, J. P.: The Influence of
684	Chronic Ozone Exposure on Global Carbon and Water Cycles, J. Climate., 28,
685	292–305, https://doi.org/10.1175/JCLI-D-14-00223.1, 2015.
686	van Marle, M. J. E., Kloster, S., Magi, B. I., Marlon, J. R., Daniau, AL., Field, R. D.,
687	Arneth, A., Forrest, M., Hantson, S., Kehrwald, N. M., Knorr, W., Lasslop, G., Li,
688	F., Mangeon, S., Yue, C., Kaiser, J. W., and van der Werf, G. R.: Historic global
689	biomass burning emissions for CMIP6 (BB4CMIP) based on merging satellite
690	observations with proxies and fire models (1750–2015), Geosci. Model. Dev., 10,
691	3329–3357, https://doi.org/10.5194/gmd-10-3329-2017, 2017.
692	Menon, S. and Rotstayn, L.: The radiative influence of aerosol effects on liquid-phase
693	cumulus and stratiform clouds based on sensitivity studies with two climate
694	models, Clim. Dyn., 27, 345–356, https://doi.org/10.1007/s00382-006-0139-3,
695	2006.
696	Mills, G., Buse, A., Gimeno, B., Bermejo, V., Holland, M., Emberson, L., and Pleijel,
697	H.: A synthesis of AOT40-based response functions and critical levels of ozone
698	for agricultural and horticultural crops, Atmos. Environ., 41, 2630–2643,
699	https://doi.org/10.1016/j.atmosenv.2006.11.016, 2007.
700 701 702 703	Mills, G., Harmens, H., Wagg, S., Sharps, K., Hayes, F., Fowler, D., Sutton, M. andDavies, B.: Ozone impacts on vegetation in a nitrogen enriched and changingclimate,Environ.Pollut.,208,898-908,https://doi.org/10.1016/j.envpol.2015.09.038, 2016.
704 705	Myhre, G., Shindell, D., Breion, FM., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, JF., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens,

706 G., Takemura, T., and Zhang, H., Anthropogenic and Natural Radiative Forcing, 707 in: Climate Change 2013: The Physical Science Basis. Contribution of Working 708 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., 709 710 Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge 711 University Press, Cambridge, UK and New York, NY, USA, 2013. Norval, M., Lucas, R. M., Cullen, A. P., De Gruijl, F. R., Longstreth, J., Takizawa, Y., 712 and Van Der Leun, J. C.: The human health effects of ozone depletion and 713 interactions with climate change, Photoch. Photobio. Sci., 10, 199-225, 714 715 https://doi.org/10.1039/C0PP90044C, 2011. Nussbaum, S. and Fuhrer, J.: Difference in ozone uptake in grassland species between 716 open-top chambers and ambient air, Environ. Pollut., 109, 463-471, 717 https://doi.org/10.1016/S0269-7491(00)00049-X, 2000. 718 Nuvolone, D., Petri, D., and Voller, F.: The effects of ozone on human health, Environ. 719 720 Sci. Pollut. R., 25, 8074–8088, https://doi.org/10.1007/s11356-017-9239-3, 2018. 721 Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 722 1980-2020, Atmos. Chem. Phys., 7, 4419-4444, https://doi.org/10.5194/acp-7-723 724 4419-2007, 2007. Oleson, K. W., Lawrence, D. M., Bonan, G. B., Flanne, M. G., Kluzek, E., Lawrence, 725 P. J., Levis, S., Swenson, S. C., and Thornton, P. E.: Technical Description of 726 version 4.0 of the Community Land Model (CLM), National Center for 727 Atmospheric Research, Boulder, USA, CONCAR/TN-478+STR, 2010. 728 Oliver, R. J., Mercado, L. M., Sitch, S., Simpson, D., Medlyn, B. E., Lin, Y.-S., and 729 730 Folberth, G. A.: Large but decreasing effect of ozone on the European carbon sink, 731 Biogeosciences, 15, 4245–4269, 2018. 732 Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of 733 hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, https://doi.org/10.5194/acp-7-1961-2007, 2007. 734 Paoletti, E., De Marco, A. and Racalbuto, S.: Why should we calculate complex indices 735 of ozone exposure? Results from Mediterranean background sites. Environ. Monit. 736 737 Assess., 128, pp.19-30, https://doi.org/10.1007/s10661-006-9412-5, 2007. Pitchford, M., Malm, W., Schichtel, B., Kumar, N., Lowenthal, D., and Hand, J.: 738 Revised Algorithm for Estimating Light Extinction from IMPROVE Particle 739 Japca. J. Air. Waste. Ma., 740 Speciation Data, 57 (11), 1326-1336. https://doi.org/10.3155/1047-3289.57.11.1326, 2007. 741 Pleijel, H., Danielsson, H., Ojanperä, K., Temmerman, L. D., Högy, P., Badiani, M., 742

and Karlsson, P. E.: Relationships between ozone exposure and yield loss in
European wheat and potato—a comparison of concentration- and flux-based
exposure indices, Atmos. Environ., 38, 2259–2269,
https://doi.org/10.1016/j.atmosenv.2003.09.076, 2004.

- Pleijel, H., Danielsson, H., Emberson, L., Ashmore, M. R., and Mills, G.: Ozone risk
 assessment for agricultural crops in Europe: further development of stomatal flux
 and flux-response relationships for European wheat and potato, Atmos. Environ.,
 41, 3022–3040, https://doi.org/10.1016/j.atmosenv.2006.12.002, 2007.
- Remer, L. A., Kaufman, Y. J., Tanré, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R.R., Ichoku, C., Levy, R. C., and Kleidman, R. G.: The MODIS aerosol algorithm,
 products, and validation, J. Atmos. Sci., 62, 947–973,
 https://doi.org/10.1175/JAS3385.1, 2005.
- Sadiq, M., Tai, A. P., Lombardozzi, D., and Val Martin, M.: Effects of ozone-vegetation
 coupling on surface ozone air quality via biogeochemical and meteorological
 feedbacks, Atmos. Chem. Phys., 17, 3055–3066, https://doi.org/10.5194/acp-173055-2017, 2017.
- Schmidt, G. A., Ruedy, R., Hansen, J. E., Aleinov, I., Bell, N., Bauer, M., Bauer, S., 759 760 Cairns, B., Canuto, V., Cheng, Y., Genio, A. D., Faluvegi, G., Friend, A. D., Hall, 761 T. M., Hu, Y., Kelley, M., Kiang, N. Y., Koch, D., Lacis, A. A., Lerner, J., Lo, K. 762 K., Miller, R. L., Nazarenko, L., Oinas, V., Perlwitz, J., Perlwitz, J., Rind, D., Romanou, A., Russell, G. L., Sato, M., Shindell, D. T., Stone, P. H., Sun, S., 763 764 Tausnev, N., Thresher, D., and Yao, M.-S.: Present-Day Atmospheric Simulations Using GISS ModelE: Comparison to In Situ, Satellite, and Reanalysis Data, J. 765 Climate., 19, 153–192, https://doi.org/10.1175/JCLI3612.1, 2006. 766
- 767 Schmidt, G. A., Kelley, M., Nazarenko, L., Ruedy, R., Russell, G. L., Aleinov, I., Bauer, 768 M., Bauer, S. E., Bhat, M. K., Bleck, R., Canuto, V., Chen, Y.-H., Cheng, Y., Clune, 769 T. L., Del Genio, A., de Fainchtein, R., Faluvegi, G., Hansen, J. E., Healy, R. J., 770 Kiang, N. Y., Koch, D., Lacis, A. A., LeGrande, A. N., Lerner, J., Lo, K. K., 771 Matthews, E. E., Menon, S., Miller, R. L., Oinas, V., Oloso, A. O., Perlwitz, J. P., 772 Puma, M. J., Putman, W. M., Rind, D., Romanou, A., Sato, M., Shindell, D. T., 773 Sun, S., Syed, R. A., Tausnev, N., Tsigaridis, K., Unger, N., Voulgarakis, A., Yao, M.-S., and Zhang, J.: Configuration and assessment of the GISS ModelE2 774 contributions to the CMIP5 archive: GISS MODEL-E2 CMIP5 SIMULATIONS, 775 776 J. Adv. Model. Earth Syst., 6, 141-184, https://doi.org/10.1002/2013MS000265, 777 2014.
- Sicard, P., De Marco, A., Dalstein-Richier, L., Tagliaferro, F., Renou, C., Paoletti, Elena,
 2016. An epidemiological assessment of stomatal ozone flux-based critical levels
 for visible ozone injury in southern European forests. Sci. Total. Environ., 541,
 729-741.

- Sitch, S., Cox, P. M., Collins, W. J., and Huntingford, C.: Indirect radiative forcing of
 climate change through ozone effects on the land-carbon sink, Nature, 448, 791–
 794, https://doi.org/10.1038/nature06059, 2007.
- Sofen, E. D., Bowdalo, D., Evans, M. J., Apadula, F., Bonasoni, P., Cupeiro, M., Ellul,
 R., Galbally, I. E., Girgzdiene, R., Luppo, S., Mimouni, M., Nahas, A. C., Saliba,
 M., and Tørseth, K.: Gridded global surface ozone metrics for atmospheric
 chemistry model evaluation, Earth Syst. Sci. Data, 8, 41–59,
 https://doi.org/10.5194/essd-8-41-2016, 2016.
- Unger, N., Zheng, Y., Yue, X., and Harper, K. L.: Mitigation of ozone damage to the
 world's land ecosystems by source sector, Nat. Clim. Chang., 10, 134–137,
 https://doi.org/10.1038/s41558-019-0678-3, 2020.
- VanLoocke, A., Betzelberger, A. M., Ainsworth, E. A., and Bernacchi, C. J.: Rising
 ozone concentrations decrease soybean evapotranspiration and water use
 efficiency whilst increasing canopy temperature, New. Phytol., 195, 164–171,
 https://doi.org/10.1111/j.1469-8137.2012.04152.x, 2012.
- van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y., Rogers,
 B. M., Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J., Yokelson, R. J.,
 and Kasibhatla, P. S.: Global fire emissions estimates during 1997–2016, Earth.
 Syst. Sci. Data, 9, 697–720, https://doi.org/10.5194/essd-9-697-2017, 2017.
- Wang, Y., Shen, L., Wu, S., Mickley, L., He, J. and Hao, J.: Sensitivity of surface ozone
 over China to 2000–2050 global changes of climate and emissions, Atmos.
 Environ, 75, 374-382, https://doi.org/10.1016/j.atmosenv.2013.04.045, 2013.
- Wesely, M. L. and Hicks, B. B.: A review of the current status of knowledge on dry
 deposition, Atmos. Environ., 34, 2261–2282, https://doi.org/10.1016/S13522310(99)00467-7, 2000.
- Wild, M., Folini, D., Schär, C., Loeb, N., Dutton, E. G., and König-Langlo, G.: The
 global energy balance from a surface perspective, Clim. Dyn., 40, 3107–3134,
 https://doi.org/10.1007/s00382-012-1569-8, 2013.
- Yue, X., Liao, H., Wang, H. J., Li, S. L., and Tang, J. P.: Role of sea surface temperature
 responses in simulation of the climatic effect of mineral dust aerosol, Atmos.
 Chem. Phys., 11, 6049-6062, https://doi.org/10.5194/acp-11-6049-2011, 2011.
- Yue, X. and Unger, N.: Ozone vegetation damage effects on gross primary productivity
 in the United States, Atmos. Chem. Phys., 14, 9137–9153,
 https://doi.org/10.5194/acp-14-9137-2014, 2014.
- Yue, X. and Unger, N.: The Yale Interactive terrestrial Biosphere model version 1.0:
 description, evaluation and implementation into NASA GISS ModelE2, Geosci.
 Model Dev., 8, 2399–2417, https://doi.org/10.5194/gmd-8-2399-2015, 2015.

- Yue, X. and Unger, N.: Fire air pollution reduces global terrestrial productivity, Nat.
 Commun., 9, 5413, https://doi.org/10.1038/s41467-018-07921-4, 2018.
- Yue, X., Keenan, T. F., Munger, W., and Unger, N.: Limited effect of ozone reductions
 on the 20-year photosynthesis trend at Harvard forest, Glob. Change Biol., 22,
 3750–3759, https://doi.org/10.1111/gcb.13300, 2016.
- Yue, X., Liao, H., Wang, H., Zhang, T., Unger, N., Sitch, S., Feng, Z., and Yang, J.:
 Pathway dependence of ecosystem responses in China to 1.5°C global warming,
 Atmos. Chem. Phys., 20, 2353–2366, https://doi.org/10.5194/acp-20-2353-2020,
 2020.
- Zhang, L., Vet, R., Brook, J. R., and Legge, A. H.: Factors affecting stomatal uptake of
 ozone by different canopies and a comparison between dose and exposure, Sci.
 Total Environ., 370, 117–132, https://doi.org/10.1016/j.scitotenv.2006.06.004,
 2006.
- Zhou, X., Yue, X., and Tian, C.: Responses of Ecosystem Productivity to Anthropogenic
 Ozone and Aerosols at the 2060, Earths Future, 12, e2023EF003781,
 https://doi.org/10.1029/2023EF003781, 2024.
- Zhu, J., Tai, A. P. K., and Hung Lam Yim, S.: Effects of ozone-vegetation interactions
 on meteorology and air quality in China using a two-way coupled landatmosphere model, Atmos. Chem. Phys., 22, 765–782,
 https://doi.org/10.5194/acp-22-765-2022, 2022.