# Response to Comments of Reviewer #2

**Manuscript number:** acp-2019-935
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**Title:** Ozone-vegetation feedback through dry deposition and isoprene emissions in a global chemistry-carbon-climate model

*This study considers the impacts on surface ozone concentrations due to two ozone vegetation feedback mechanisms, the dry deposition inhibition by ozone and the isoprene emission inhibition by ozone. This is an important scientific question that have been tackled by several previous studies. The unique aspect of this work is that the two feedback mechanisms are explicitly included in the ModelE2-YIBs model, and two levels of parameterized sensitivity were assessed for each of the two feedback mechanisms. The results show that the ozone-inhibition of dry deposition generally wins over the effects of ozone-inhibition of isoprene emissions, such that surface ozone increase over Eastern US, Europe, and Eastern China when the ozone effects are considered, relative to the control simulation (where no ozone effects are considered). In addition, indirect impacts on meteorology via weakened transpiration and enhanced solar radiation scattering by SOA also play a role.*

*Overall, I have a very favorable impression of this conceptual paper and consider it publishable after minor revisions. I do wish, however, that the authors can go beyond the common model validation methods and try to validate the model performance on the ozone-vegetation sensitivity. There are also key details about the model setup that needs to be included in the manuscript. See the comments below.*

**Response:**

Thank you for the helpful comments and suggestions. We have revised the manuscript carefully and the point-to-point responses are listed below.

*Major comments:*

*Section 2.1: What oxidants were considered from the two-product SOA production scheme? If ozone is one of the oxidants considered, is there significant feedback through this pathway (more O3 -> more SOA -> cooling -> reduced isoprene emission) ? The pathway that the authors described was (more isoprene -> more SOA -> cooling -> reduced isoprene emission)*

**Response:**

For the two-product SOA production scheme applied in ModelE2-YIBs, $O_3$ is the only oxidant that considered.

We further examine the feedback of 'more $O_3$ -> more SOA -> cooling'. Since $O_3$ concentrations are significantly enhanced (more $O_3$) when considering the effect of $O_3$ damage to photosynthesis and stomatal conductance (Fig. 5a and 5b), differences of SOA shortwave radiative forcing between DRY_high or DRY_low and CTRL experiments can be utilized to check whether SOA increases with more $O_3$. As is shown in Fig. R1, the SOA forcing shows very limited changes in eastern China, eastern America, and western Europe, where large $O_3$ enhancements are predicted (Fig. 5a and 5b). Such magnitude is much smaller than that in Fig. 9, which stands for the other pathway (more isoprene -> more SOA -> cooling). As a result, the weaker SOA cooling effect is driven by damaged IPE rather than the enhanced $O_3$ concentrations.

Meanwhile, we did not consider the feedback of SOA cooling on isoprene emissions. Instead, we speculated that weaker SOA cooling (less SOA) promoted temperature and surface $O_3$ concentrations in eastern U.S.

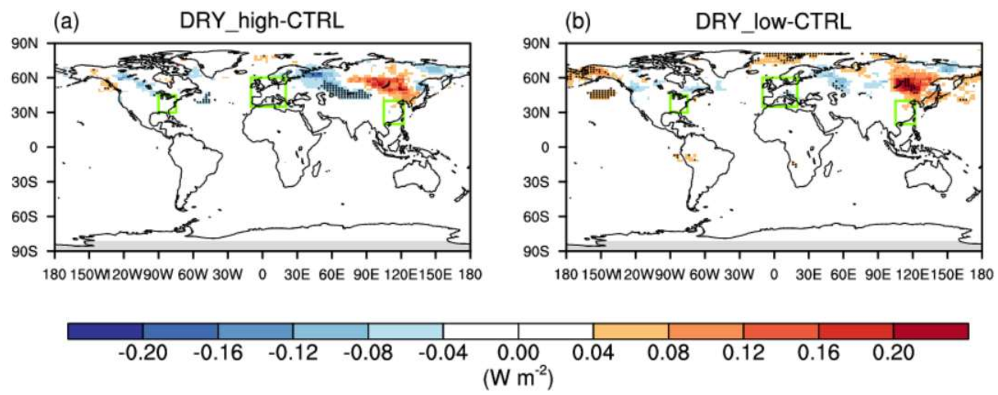



Figure R1. Effects of (a) high and (b) low $O_3$ vegetation damages on SOA shortwave radiative forcing at the surface during the boreal summer. Dotted grids indicate significant changes at the 95% confidence level. Eastern China, eastern U.S. and western Europe are enclosed by green rectangles.

*Section 2.1: What assumptions were made regarding isoprene nitrate formation and its photochemical fate? This has long been shown to significantly impact the response of ozone to isoprene emissions.*

**Response:**

Only three chemical reactions are considered in ModelE2-YIBs related to isoprene:

$$C_5H_8 + OH \rightarrow HCHO + Alkenes$$

$$C_5H_8 + O_3 \rightarrow HCHO + Alkenes$$

$$C_5H_8 + NO_3 \rightarrow HO_2 + Alkenes$$

Both HCHO and $HO_2$ further contribute to the formation of ozone.

The last paragraph of Sect. 2.1 has been revised as:

'Isoprene and α-pinene are considered as the precursors for biogenic secondary organic aerosols (SOA) in ModelE2-YIBs, which are computed online based on the two-product scheme developed by Chung and Seinfeld (2002). Isoprene can be oxidized by $O_3$ as follows:

$$C_5H_8 + O_3 \rightarrow HCHO + A_1P_1 + A_2P_2$$
(5)

Changes for semivolatile product $P_i$ (i=1,2) at each time step (*dt*) are calculated by:

$$\frac{dP_i}{dt} = A_i * rr * [O_3] * [C_5H_8]$$
(6)

where rr is the chemical reaction rate of $O_3$ and isoprene calculated by Arrhenius equation. $[O_3]$ and $[C_5H_8]$ are the $O_3$ and isoprene concentrations, respectively. $A_i$ is the molar based stoicheiometric coefficient depending on SOA formation pathways (high or low $NO_x$) (Lane et al., 2008). Temperature (T) dependence on partitioning coefficient (Kp) for P1 and P2 are given by the Clausius-Clapeyron equation:

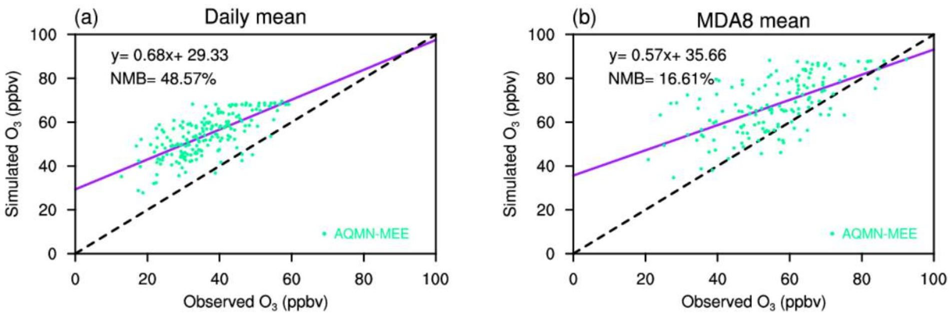
$$K_p = K_{sc}\frac{T}{T_{sc}}\exp\left[\frac{\Delta H}{R}\left(\frac{1}{T} - \frac{1}{T_{sc}}\right)\right]$$
(7)

where ΔH is the enthalpy of vaporization and is set as 42.0 kJ $mol^{-1}$ for isoprene (Chung and Seinfeld, 2002;Henze and Seinfeld, 2006) and 72.9 kJ $mol^{-1}$ for α-pinene. Ksc is the saturation concentrations at the temperature Tsc (295 K) and set as 1.62 (0.064) $m^3$ $\mu g^{-1}$ and 0.0086 (0.0026) $m^3$ $\mu g^{-1}$ for the two products formed by oxidation of isoprene (α-pinene), respectively (Presto et al., 2005;Henze and Seinfeld, 2006).' (Page5 Lines 21-31; Page 6 Lines 1-4)

*The validation of the model performance in reproducing surface ozone concentration is unsatisfactory. The model, while no worse than others, does not reproduce well the ozone observations. More importantly, validating the mean surface ozone level does not really give insights to whether the model correctly (or better than other models) reproduces the ozone-vegetation relationship. I wish the authors can make an effort to* *go the extra mile and look at the ozone-temperature dependancy or the ozoneLAI dependency. Also, does the model perform better in the sensitivity simulations including vegetation-chemistry feedbacks?*

**Response:**

Simulated $O_3$ concentrations do show certain biases compared to surface observations. However, if we validate maximum daily 8-hour average (MDA8) [$O_3$], we found that the model shows much lower biases (Fig. S1 in the revised manuscript). The main reason for the overestimation is that the model predicts high nighttime [$O_3$] that are not consistent with observations. Since $O_3$-vegetation interactions usually occur in the daytime, the updated validation shows that ModelE2 is good to use for this study.




Supplementary Fig. S1. Scatter plots of (a) daily mean and (b) MDA8 $O_3$ concentrations (ppbv) over observational sites in China. The purple line shows the linear regression between the observed and simulated $O_3$ concentrations. The black dashed line shows the 1:1 lines.

Ozone-vegetation relationships have been fully evaluated in our previous studies. For example, we validated $O_3$-GPP relations for six main vegetation types in Yue and Unger (2018) as follows:

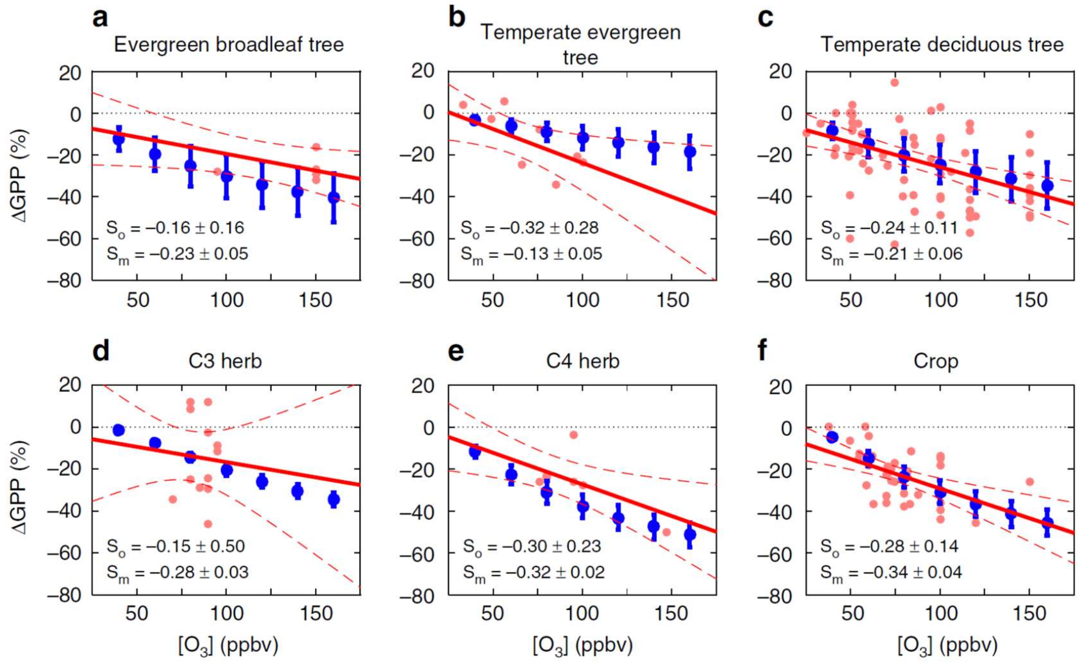



**Figure R2.** Percentage changes in GPP for six main plant functional types (PFTs) caused by $O_3$. Red points on each panel represent literature-based measurements. The linear regression is denoted as a red solid line, with 95% confidence intervals shown as dashed lines. Blue points represent simulated GPP changes from offline sensitivity experiments (Methods), with error bars indicating the range of prediction from low to high $O_3$ damaging sensitivities. The slopes of observed ($S_o$, mean ± 95% confidence interval) and modeled ($S_m$, mean ± (high-low)/2 sensitivity) GPP-$O_3$ sensitivity is shown on each panel (figure from Yue, X., and Unger, N.: Fire air pollution reduces global terrestrial productivity, Nature Communications, 9, 5413, 2018).

We validated $O_3$ damages to stomatal conductance for deciduous trees in Yue et al. (2016) as follows:

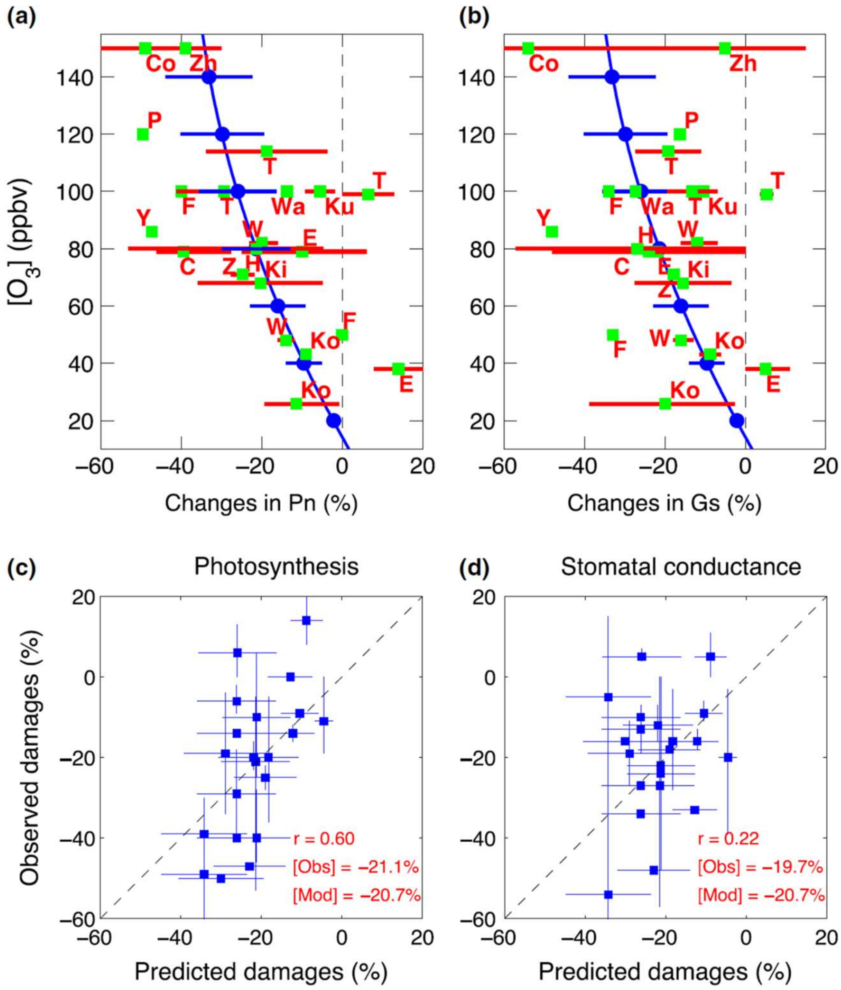



**Figure R3.** Percentage changes in (a) photosynthesis and (b) stomatal conductance averaged across 20 deciduous broadleaf forest flux tower sites in response to different levels of [$O_3$]. The derived percentage changes (including uncertainties) based on the fits are plotted against observations for (c) photosynthesis and (d) stomatal conductance (figure from Yue, X., Keenan, T. F., Munger, W., and Unger, N.: Limited effect of ozone reductions on the 20-year photosynthesis trend at Harvard forest, Global Change Biology, 22, 3750-3759, 2016).

The relations between $O_3$ and LAI can not be evaluated as such observations are not available. However, based on good performance in simulating $O_3$-GPP and GPP-LAI (Yue and Unger, 2015) relationships, we consider ModelE2-YIBs model is appropriate to use for exploring $O_3$-vegetation interactions.

Finally, inclusion of $O_3$-vegetation feedback does not necessarily improve the model performance. The main purpose for this study is to explore the processes and magnitude of $O_3$-vegetation feedback. The positive feedback we revealed may further enhance the model biases, suggesting that additional efforts are required to reduce modeling uncertainties in surface $O_3$.

*The authors suggested that the reason for over-estimation of ozone over China was due to an overestimation of anthropogenic emissions? Is there justification for that? How does the IPCC RCP8.5 emission (van Vuuren et al., 2011) compare to Chinese inventories. The authors also did not mention the basis of their isoprene emission. Have the authors validated their isoprene emissions for the three regions against inversion studies using satellite observations?*

**Response:**

We have revised the first paragraph in Sect. 3.1 as follow:

'Figure 1 shows a comparison of the simulated summer $O_3$ concentrations to the observations. The model in general captures reasonable spatial patterns with a correlation coefficient of 0.41. The NMBs between simulations and observations in U.S and Europe are 11.7% and 13.2%, respectively, which are comparable with the simulation performed by CESM (Lamarque et al., 2012; Sadiq et al., 2017). However, the model overestimates $O_3$ concentrations by 29.3% with a regression intercept of 32 ppbv, suggesting that simulated $O_3$ vegetation damage might be overestimated especially over some regions with low ambient $O_3$ level. The large overestimate is mainly a result of overestimation in China. However, if we validate maximum daily 8-hour average (MDA8) $O_3$ concentrations, we found that the model shows much lower biases (Fig. S1). The main reason for the overestimation is that the model predicts high nighttime $O_3$ concentrations that are not consistent with observations. Since $O_3$-vegetation interactions usually occur in the daytime, the validation shows that ModelE2-YIBs is good to use for this study. Meanwhile, most of the observational sites in AQMN-MEE are located in urban area, which might be another reason for the surface $O_3$ overestimates in China (Yue et al., 2017).' (Page 8, Lines 17-27)

As for the isoprene emissions, extensive validation has been done in previous study (Unger et al., 2013). They showed that a control simulation reproduced 50% of the variability across different ecosystems and seasons in a global database of 28 measured campaign-average fluxes, and captured the observed variance in the 30 min average diurnal cycle ($R^2$ =64–96%) at nine sites. The description of isoprene emissions has been added in the second paragraph in Sect. 2.1:

‘…The LAI and tree growth are dynamically simulated with the allocation of carbon assimilation. The emissions of isoprene are calculated online as a function of Je photosynthesis (Eq. 1), canopy temperature, intercellular $CO_2$, and $CO_2$ compensation point (Arneth et al., 2007;Unger, 2013), and have been fully validated by Unger et al. (2013). Carbon fluxes, phenology, LAI, GPP, and net ecosystem exchange (NEE), ….’ (Page 5, Lines 1-5)

*Minor comments:*

*Page 4, Lines 23-25: What is the scientific basis for parameterizing stomatal conductance as a function of these parameters, especially A_tot? I realize that a full answer to this question is beyond the scope of this study. Nevertheless, it might worthwhile to say a few words here or in the introduction to justify this assumption, which is central to the results of this study.*

**Response:**

Plant photosynthesis is closely related to stomatal conductance. The higher A_tot requires larger Gs to allow more $CO_2$ enter the leaves for photosynthesis. Such relationship has been parameterized by the Farquhar and Ball-Berry models, which has been widely utilized in land ecosystem simulation (e.g. Farquhar et al., 1980;Ball et al., 1987;Sitch et al., 2007;Bonan et al., 2011;Lombardozzi et al., 2012;Yue and Unger, 2015;Deryng et al., 2016;Sadiq et al., 2017).

The second paragraph of Sect. 2.1 has been revised as:

‘The YIBs model is a dynamic vegetation model that includes 9 plant functional types (PFTs) (Table S1) and can simulate biophysical processes of photosynthesis, transpiration and respiration with variations in meteorological fields. **Since the higher leaf photosynthesis requires larger stomatal conductance to allow more $CO_2$ enter the leaves, leaf photosynthesis and stomatal conductance are closely related** and calculated using the Farquhar and Ball–Berry models **(Farquhar et al., 1980;Ball et al., 1987)** as follows:

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

$$g_s = m\frac{(A_{tot} - R_d) \times RH}{c_s} + b \quad (2)$$

where the total leaf photosynthesis ($A_{tot}$) is the minimum value of the ribulose-1,5-bisphosphate carboxylase (RuBisCO)-limited rate of carboxylation ($J_c$), light-limited rate ($J_e$), and export-limited rate ($J_s$). **Stomatal conductance for $H_2O$ ($g_s$)** is calculated by the $A_{tot}$, dark respiration rate ($R_d$), relative humidity ($RH$) and $CO_2$ concentration at the leaf surface ($c_s$). The values of $m$ and $b$ are different for different PFTs (Table S1). A canopy radiation scheme is applied in YIBs to separate diffuse and direct light for sunlit and shaded leaves **(Spitters et al., 1986)**. The LAI and tree growth are dynamically simulated with the allocation of carbon assimilation. Carbon fluxes, phenology, LAI, GPP, and net ecosystem exchange (NEE), as well as other parameters of vegetation in ModelE2-YIBs, have been previously extensively evaluated and agree well with the observations (Yue and Unger, 2015). **In addition, ModelE2-YIBs shows good performance in simulating $O_3$-vegetation interactions such as $O_3$-GPP and $O_3$-$g_s$ relationships (Yue et al., 2016; Yue et al., 2018).**' (Page 4 Lines 21-31; Page 5 Lines 1-7)

*Page 4, Lines 25-26: missing reference for the canopy radiation scheme.*

**Response:**

Revised.

*Page 5, line 12: 'online computed' should be 'computed online'*

**Response:**

Revised.

*Page 5, line 27: How was F_O3 calculated and how was it related to g_s?*

**Response:**

The equation for $F_{O3}$ calculation has been added as follow:

'A semi-mechanistic scheme proposed by Sitch et al. (2007) is applied in this study that simulates the effect of $O_3$ damage to the photosynthesis rate and stomatal conductance via the following formulas:

$$\mathrm{A_{totd}} = \mathrm{F} \times \mathrm{A_{tot}} \quad (8)$$

$$g_{sd} = F \times g_s \quad (9)$$

where $A_{totd}$ ($g_{sd}$) and $A_{tot}$ ($g_s$) are the $O_3$-affected and original total leaf photosynthesis (stomatal conductance), respectively. $F$ is the ratio between affected and original photosynthesis. It depends on the instantaneous leaf uptake of $O_3$ as follows:

$$F = 1 - a \times max\,[F_{O_3} - F_{O_3,crit}, 0.0] \quad (10)$$

where parameter $a$ represents the $O_3$ damaging sensitivity dependent on vegetation types with a range from low to high values. $F_{O3,crit}$ is a critical threshold for damage (Table S1). $F_{O3}$ is the $O_3$ uptake rate by the stomata, which is calculated by:

$$F_{O3} = \frac{[O_3]}{R_a + [\frac{k_{O3}}{g_{sd}}]} \quad (11)$$

Where $[O_3]$ is the surface $O_3$ concentrations and $R_a$ is the aerodynamic resistance in Eq. (3). $k_{O3}$ is 1.67, which is the ratio of leaf resistance for $O_3$ to leaf resistance for water vapor. This scheme has been used to explore $O_3$ damages to vegetation in many previous studies…..' (Page 6 Lines 7-19)

*Page 6, line 23: What is n in Equation (10)?*

**Response:**

The Eq. (10) in the origin manuscript and the correspondingly explanation has been revised as:

$$POD_1 = \int_1^n (F_{O_3} - 1)dt$$

(14)

'where $F_{O3}$ is the $O_3$ uptake rate by stomata (nmol $O_3$ $m^{-2}$ $s^{-1}$), which is the same as that in Eq. (11). *dt* indicates the time integration step and *n* indicates the total number of time steps during the growing season.' (Page 7 Lines 10-12)

*Figure 1b: Please label the x and y axes. Also, the pastel colors in Figures 1b and S1 are extremely hard to see. Please consider changing the color scheme.*

**Response:**

Revised.

Figure 1:

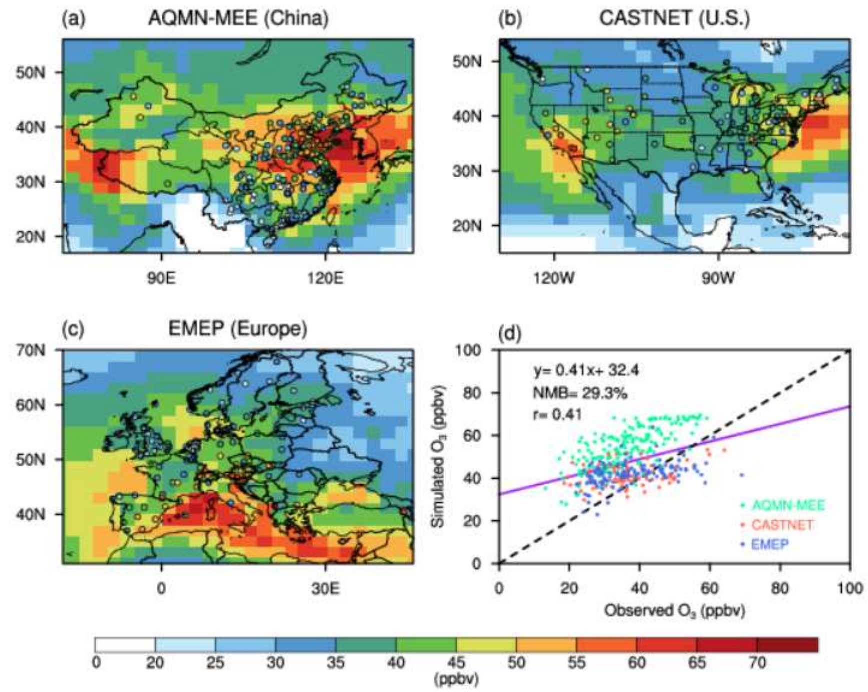
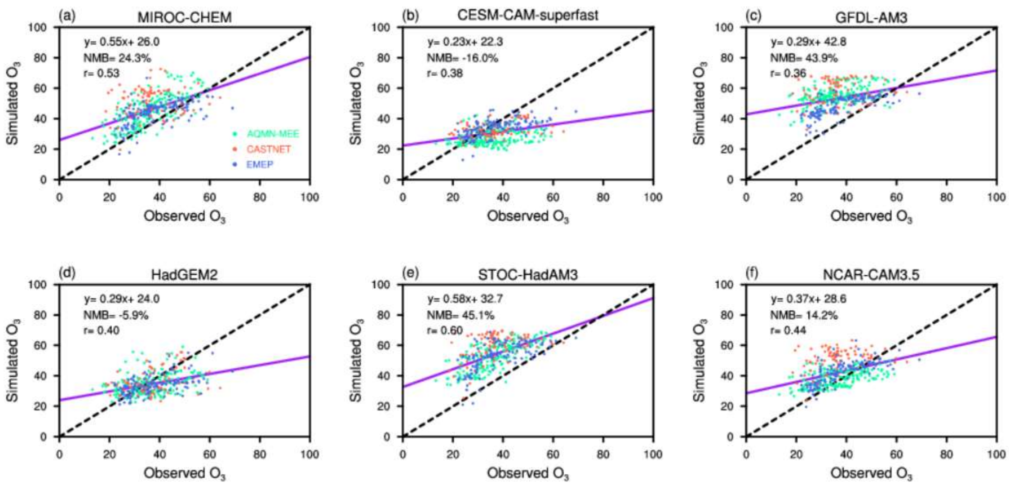



Figure S1 (Figure S2 in the revised manuscript):



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