**Referee 1: Prof. William Collins**

We are grateful to Prof. William Collins for his time and energy in providing helpful comments and guidance that have improved the manuscript. In this document, we describe how we have addressed the reviewer's comments. Referee comments are shown in black italics and author responses are shown in blue regular text.

*The paper by Yue et al. is a valuable assessment of effects of ozone and aerosol pollution on NPP over China. In particular this is the first time the aerosol contribution has been examined in such detail. The modelled ozone damage is compared against field measurements. This paper should certainly be published in ACP, however some revision is needed as described below.*

*The authors should make it clearer how much of the impact of ozone and aerosols on NPP is natural and how much anthropogenic. The headline numbers are referred to as due to air pollution, but presumably there would be effects on NPP due to natural ozone. Two extra runs G10NATLO3 and G10NATHO3 would provide the required data for this. The authors assert that since the NPP effect is small below 40 ppb the no ozone and natural ozone simulations are equivalent, but the authors need to demonstrate this with these two extra runs.*

→ We performed two extra runs, G10NATLO3_OFF and G10NATHO3_OFF, which consider offline vegetation damages due to $O_3$ from natural emissions alone. We added a new Figure S7 to compare the GPP reductions caused by $O_3$ with and without anthropogenic emissions. It shows that $O_3$ from natural sources has trivial impacts on GPP.

In the Methods section, we revised the text to introduce the two additional experiments: "To compare the differences between online and offline $O_3$ damage, we perform <u>four additional simulations</u> which do not account for the feedbacks of $O_3$-induced changes in biometeorology, plant growth, and ecosystem physiology. Two simulations, G10ALLHO3_OFF and G10ALLLO3_OFF, include both natural and anthropogenic emissions. <u>The other two, G10NATHO3_OFF and G10NATLO3_OFF, include natural emissions alone.</u>" (Lines 253-259)

In the Results section, we describe the findings from the extra runs: "Sensitivity simulations with zero anthropogenic emissions show almost no $O_3$ damage (Fig. S7), because the $[O_3]$ exposure from natural sources alone is usually lower than the threshold level of 40 ppbv below which the damage for most PFTs is limited (Fig. 5)." (Lines 402-405)

*When analysing the meteorological changes the authors only show the impacts over China. In their global model set up the aerosols will change globally and affect global circulation (even with fixed SST). The authors should therefore show global maps corresponding to figures 7 and S8 in the supplement. One feature of perturbing aerosols in fixed-SST simulations is that there are large changes in the land-sea temperature* *contrast and hence artificial changes in circulation patterns. The resulting meteorological changes over China will therefore be a combination of locally driven effects (such as change in radiation and hence evaporation) and regional-globally driven effects (such as changes in rainfall and hence soil water). This seems to be particularly apparent in the AIE simulations where the patterns of changes in precipitation and soil water bear no relation to the changes in aerosol. The soil moisture changes dominate the aerosol impacts on NPP and I am not convinced these can be attributed to the aerosol changes. The changes in PAR and surface temperature can be much more readily linked physically to the changes in aerosol, therefore the authors should exclude the soil moisture changes from their analysis in table 2.*

We do agree that the aerosol-induced changes in meteorology over China are a combination of local and remote effects, but we assert that the changes (local and remote) are all ultimately attributed to the aerosol radiative perturbation. For example, the regional soil moisture changes are absolutely caused by the aerosol radiative perturbations, but may be more linked to a regionally-globally driven dynamical mechanism rather than the reduction in local downward shortwave. The main goal of this study is to investigate the terrestrial biospheric response to air pollution in China. Diagnosing long range dynamical mechanisms is out of scope of this study, especially because the aerosol impacts are so much smaller than the ozone impacts, this specific study will not gain from an explicit description of the multi-scale dynamical mechanisms that drive the regional meteorological changes. Furthermore, the paper is already quite long with 10 figures and supporting information, and global maps obscure and make it difficult for readers to see the regional signals in China. Therefore, we decide not to add further figures of global maps corresponding to figures 7 and S9 in the supplement. In Section 4.1, we already include a comparison of the simulated aerosol impacts on meteorology and surface climate against existing published estimates and a discussion of their realism and uncertainties. Similarly, we select to retain the soil moisture results in Table 3 (original Table 2). Soil moisture dominates the NPP response in only 2 out of 6 scenarios/cases. Even if the soil wetness changes occur in part through more long range dynamical mechanisms triggered by the aerosol radiative perturbations, it is important to highlight the biospheric sensitivity to changes in this driver, oftentimes ignored/neglected in aerosol-carbon-climate studies. For example, precipitation controls GPP in more than 40% of vegetated land (Beer et al., Science, 2010).

Following Prof. Collin's suggestions, we make several modifications.

We add: "The resulting meteorological changes over China are a combination of locally driven effects (such as changes in radiation and hence temperature) and regional-globally driven effects (such as changes in rainfall and hence soil water)." (Lines 431-434).

We clarify: "Aerosol-induced impacts on precipitation and soil moisture are not statistically significant over the regionally averaged domain (Tables S5 and S6). However, for the 2010 and 2030 CLE cases with AIE, 2 out of 6 scenarios, the aerosol-induced impact on soil moisture dominates the total NPP response (Table 3)." (Lines 626-629)

We emphasize: "our estimate of NPP response to aerosol effects, with or without AIE, is secondary in magnitude compared to the $O_3$ vegetation damage." (Lines 633-634)

*More explanation of table 2 is needed. What simulations are compared against what to derive the answers? How are the uncertainties derived? – presumably they are interannual variability, but different sets of annually-varying data are used for the online and offline calculations.*

→ We added more descriptions in the footnote of Table 3 (original Table 2) to explain how we derive those numbers from different simulations. We clarified in the section 2.4.1 about the uncertainties: "The full list of simulations in Table 2 offers assessment of uncertainties due to interannual climate variability, emission inventories (CLE or MTFR), $O_3$ damage sensitivities (low to high), and aerosol climatic effects (direct and indirect). Uncertainties calculated based on the interannual climate variability in the model are indicated using the format 'mean ± one standard deviation'. Other sources of uncertainty are explicitly stated." (Lines 276-281).

The interannual climate variability from online and offline simulations are calculated using different time periods. We clarified in the section 2.4.3 as follows: "Uncertainties due to interannual climate variability in the model are calculated using different time periods for the online (15 years, Table 2) and offline (10 years, Table S3) runs." (Lines 311-313).

***Specific points***

*Page 2, lines 47-53: Uncertainties (including the range between high and low sensitivity) need to be included here.*

→ We included uncertainties due to $O_3$ sensitivity: "In the present day, $O_3$ reduces annual NPP by 0.6 Pg C (14%) with a range from 0.4 Pg C (low $O_3$ sensitivity) to 0.8 Pg C (high $O_3$ sensitivity). In contrast, aerosol direct effects increase NPP by 0.2 Pg C (5%) through the combination of diffuse radiation fertilization, reduced canopy temperatures, and reduced evaporation leading to higher soil moisture. Consequently, the net effects of $O_3$ and aerosols decrease NPP by 0.4 Pg C (9%) with a range from 0.2 Pg C (low $O_3$ sensitivity) to 0.6 Pg C (high $O_3$ sensitivity). However, precipitation inhibition from combined aerosol direct and indirect effects reduces annual NPP by 0.2 Pg C (4%), leading to a net air pollution suppression of 0.8 Pg C (16%) with a range from 0.6 Pg C (low $O_3$ sensitivity) to 1.0 Pg C (high $O_3$ sensitivity)." (Lines 47-56)

*Page 2, line 55: suggest to replace "will not alleviate" with "will be further increased".*

→ Corrected as suggested.

*Page 4, line 93: "not been properly validated" – The authors need to be more explicit about exactly what deficiencies the previous studies had.*

→ We clarified the sentence as follows: "Previous regional modeling … does not always take advantage of valuable observations to calibrate GPP-$O_3$ sensitivity coefficients for the China domain and typically the derived results have not been properly validated." (Lines 94-97)

*Page 7, line 186: The authors should clarify that they are referring to 2010 emissions.*

→ We clarified as follows: "Inter-comparison of present-day (the year 2010) emissions (Fig. S2) shows …" (Lines 190-191)

*Page 7, line 201: How much does biomass burning contribute to the emissions? Are they considered natural (in G10NATxxx)?*

→ We clarified as follows: "Biomass burning emissions, adopted from the IPCC RCP8.5 scenario (van Vuuren et al., 2011), are considered as anthropogenic sources because most fire activities in China are due to human-managed prescribed burning. Compared with the GAINs inventory, present-day biomass burning is equivalent to <1% of the emissions for $NO_x$, $SO_2$, and $NH_3$, 1.6% for BC, 3.0% for CO, and 9.6% for OC." (Lines 213-217)

*Page 7, lines 205-210: The authors need to describe how the changes in natural emissions are determined.*

→ We add a detailed description of the climate-sensitive natural emission sources:

"The model represents climate-sensitive natural precursor emissions of lightning $NO_x$, soil $NO_x$ and biogenic volatile organic compounds (BVOCs) (Unger and Yue, 2014). Future 2030 changes in these natural emissions are small compared to the anthropogenic emission changes. Interactive lightning $NO_x$ emissions are calculated based on the climate model's moist convection scheme that is used to derive the total lightning and the cloud-to-ground lightning frequencies (Price et al., 1997; Pickering et al., 1998; Shindell et al., 2013). Annual average lightning $NO_x$ emissions over China increase by 4% between 2010 and 2030. Interactive biogenic soil $NO_x$ emission is parameterized as a function of PFT-type, soil temperature, precipitation (including pulsing events), fertilizer loss, LAI, $NO_x$ dry deposition rate, and canopy wind speed (Yienger and Levy, 1995). Annual average biogenic soil $NO_x$ emissions increase by only 1% over China between 2010 and 2030. Leaf isoprene emissions are simulated using a biochemical model that depends on the electron transport-limited photosynthetic rate, intercellular $CO_2$, canopy temperature, and atmospheric $CO_2$ (Unger et al., 2013). Leaf monoterpene emissions depend on canopy temperature and atmospheric $CO_2$ (Unger and Yue, 2014). Annual average isoprene emission in China increases by 5% (0.39 TgC/yr) between 2010 and 2030 in response to enhanced GPP and temperature that offset the effects of $CO_2$-inhibition. Monoterpene emissions decrease by 5% (-0.25 Tg C) between 2010 and 2030 because $CO_2$-inhibition outweighs the effects of increased warming." (Lines 220-238).

*Page 8, line 217. I suggest including the table of simulations in the main text rather than the supplement.*

→ We have moved Table S2 into the main text as suggested (now Table 2).

*Page 8, line 232. The authors should list the changes in WMGHG from 2010 to 2030 (at least CO2 and methane).*

→ We clarified as follows: "Well-mixed GHG concentrations are also adopted from the RCP8.5 scenario, with $CO_2$ changes from 390 ppm in 2010 to 449 ppm in 2030, and $CH_4$ changes from 1.779 ppm to 2.132 ppm." (Lines 264-266)

*Page, 9 section 2.4.3. This section isn't clear about how the meteorological changes are applied to the offline model. Are they applied as an average (of the last 15 years) of the table S2 simulations; or are individual years from the table S2 simulations used as input. If the former: why is there any variability in the offline output? If the latter: the last 3 years could be strongly influenced by interannual variability.*

→ We add one sentence to explain how the meteorological changes are applied to the offline model: "For these simulations, the month-to-month meteorological perturbations caused by aerosols are applied as scaling factors on the baseline forcing." (Lines 307-309) We actually run the offline simulations for 15 years, with the last 10 years used for the analyses. The paper has been updated for several versions. In the first version, we ran the offline simulations for only 10 years and did not show the uncertainties in the Table because the time period was short. In the latest version, we have already re-ran all simulations to 15 years and calculated uncertainties for Table 3. However, we had omitted to update the corresponding manuscript text.

*Page 10, lines 298-299. The agreement in figure 3 doesn't suggest that the "Evaluations at rural sites better match the observations". The correlation is no better than for all sites, and by eye only the summer points look to show any correlation at all.*

→ We revise the text as follows: "Evaluations at rural sites (Table S4), which represent the major domain of China, show a mean bias of -5% (Fig. 3). The magnitude of such bias is much lower than the value of 42.5% for the comparisons at urban-dominant sites (Fig. 2f)." (Lines 346-348)

*Page 12, line 346-348. The online model presumably allows the g_s changes to feed back on the ozone concentrations, which should increase them. Therefore it might be expected that the online model would show more ozone damage. The authors should compare the g_s and surface ozone concentration changes between the online and offline models.*

→ The online model does allow $g_s$ changes to feed back onto the atmospheric composition. However, our model does not show significant $O_3$ concentration feedbacks at the current level of vegetation and $g_s$ damage, likely because of multiple offsetting influences on chemistry. We have now included $g_s$ changes in a revised Figure S6 and described them in the text as follows: "At the same time, the $O_3$-induced reductions in stomatal conductance (Fig. S6a) can increase canopy temperature and inhibit plant transpiration, leading to surface warming (Fig. S6b), dry air (Fig. S6c), and rainfall deficit (Fig. S6d)." (Lines 395-398)

*Page 12, lines 348-349. Are the authors saying they have carried out a G10NATLO3 simulation, and the NPP change (compared to G10NATNO3) is identically zero everywhere? If so, this needs to be explained more clearly. If not, then the authors need to be clearer about the evidence they have that the zero anthropogenic emissions show no damage. The damage functions in fig 5 don't go exactly to zero at 40 ppb.*

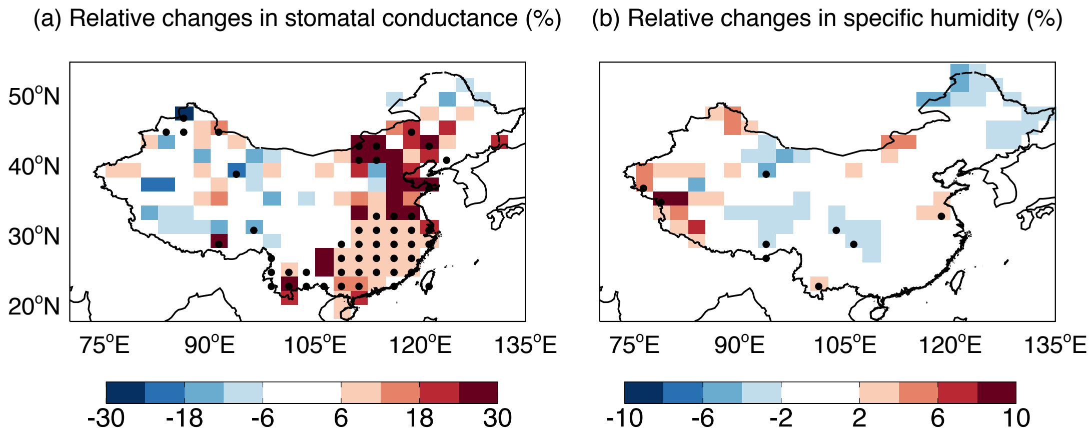
→ We performed two additional sensitivity runs with the climate model ModelE2-YIBs, G10NATLO3_OFF and G10NATHO3_OFF, to examine damages by natural $O_3$. We found negligible instead of zero impacts of background $O_3$ from natural precursor sources in China. We show the results in the new Figure S7. In the main text, we clarified as follows: "Sensitivity simulations with zero anthropogenic emissions show almost no $O_3$ damage (Fig. S7), because the [O3] exposure from natural sources alone is usually lower than the threshold level of 40 ppbv below which the damage for most PFTs is limited (Fig. 5)." (Lines 402-405)

*Page 12, line 352. The uncertainty here also needs to include the uncertainty in plant sensitivity (i.e. the range from high to low). Technically you should refer to the "central value" between high and low, rather than "average".*

→ We clarified as follows: "Our results indicate that present-day surface $O_3$ causes strong inhibitions on total NPP in China, ranging from 0.43 ± 0.12 Pg C $yr^{-1}$ with low sensitivity to 0.76 ± 0.15 Pg C $yr^{-1}$ with high sensitivity (Table 3). The central value of NPP reduction by $O_3$ is 0.59 ± 0.11 Pg C $yr^{-1}$, assuming no direct impacts of $O_3$ on plant respiration." (Lines 405-409)

*Page 12, line 362. Have the authors checked whether the absolute relative humidity is affected, i.e. whether the relative humidity change is purely due to the decreased temperature.*

→ We plotted changes in stomatal conductance and specific humidity (the figure below). It shows that specific humidity changes little. As a result, most of the changes in RH are driven by lower saturation vapor pressure. In the text, we clarified as follows: "Reduced insolation decreases summer surface temperature by 0.63°C in the East, inhibiting evaporation but increasing relative humidity (RH) due to the lower saturation vapor pressure (Table S5). These feedbacks combine to stimulate photosynthesis (Fig. 8a), which, in turn, strengthens plant transpiration (not shown)." (Lines 417-421).




*Page 12, lines 363-364. There are a lot of statements presented here without any evidence. The authors have not shown strengthened plant transpiration and have not demonstrated that any increase in RH is due to this (rather than simply decreased temperatures or increased horizontal moisture transport). Similarly the authors have not shown diagnostics demonstrating a direct causal chain between transpiration and precipitation or cloud cover. Both of these could instead be due to changes in circulation patterns.*

→ We agree that the original statements were rather terse. As shown above, the increase in transpiration (left panel of the above figure) does not increase specific humidity (right panel). Advection and convergence may alter the local moisture budget. In the revised text, we clarify as follows: "Atmospheric circulation and moisture convergence are also altered due to the pollution-vegetation-climate interactions, resulting in enhanced precipitation (Fig. 7b) and cloud cover (Fig. 7d)." (Lines 421-423)

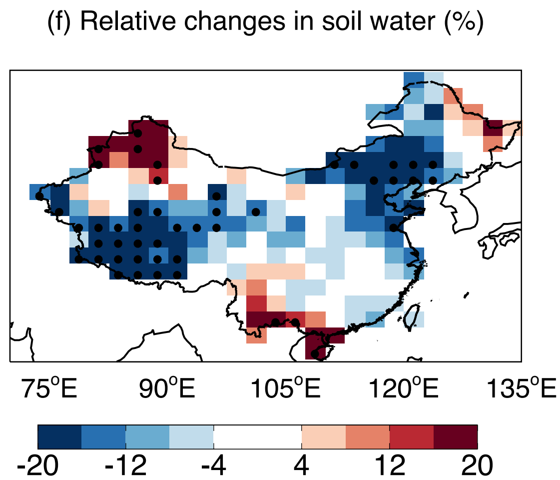
*Page 12, lines 367-369. Again no evidence is presented that the decrease in summer precipitation is due to a reduction in the cloud droplet size. Ultimately precipitation is driven by moisture convergence.*

→ The precipitation changes are due to a combination of altered cloud microphysics and atmospheric circulation patterns for which we have no way of disentangling. We modify the statement as follows: "Inclusion of AIE results in distinct climatic feedbacks (Fig. S9). Summer precipitation decreases by 0.9 mm $day^{-1}$ (13%), leading to a 3% decline in soil moisture (Table S6)." (Lines 424-426)

*Page 13, lines 382-384. This sentence wasn't very clear. Is it referring to changes in heterotrophic respiration? If so, it should be said explicitly.*

→ We have removed the sentence about the impact of phenological changes. Quantification of such effects requires additional simulations, which are out of the scope of this study.

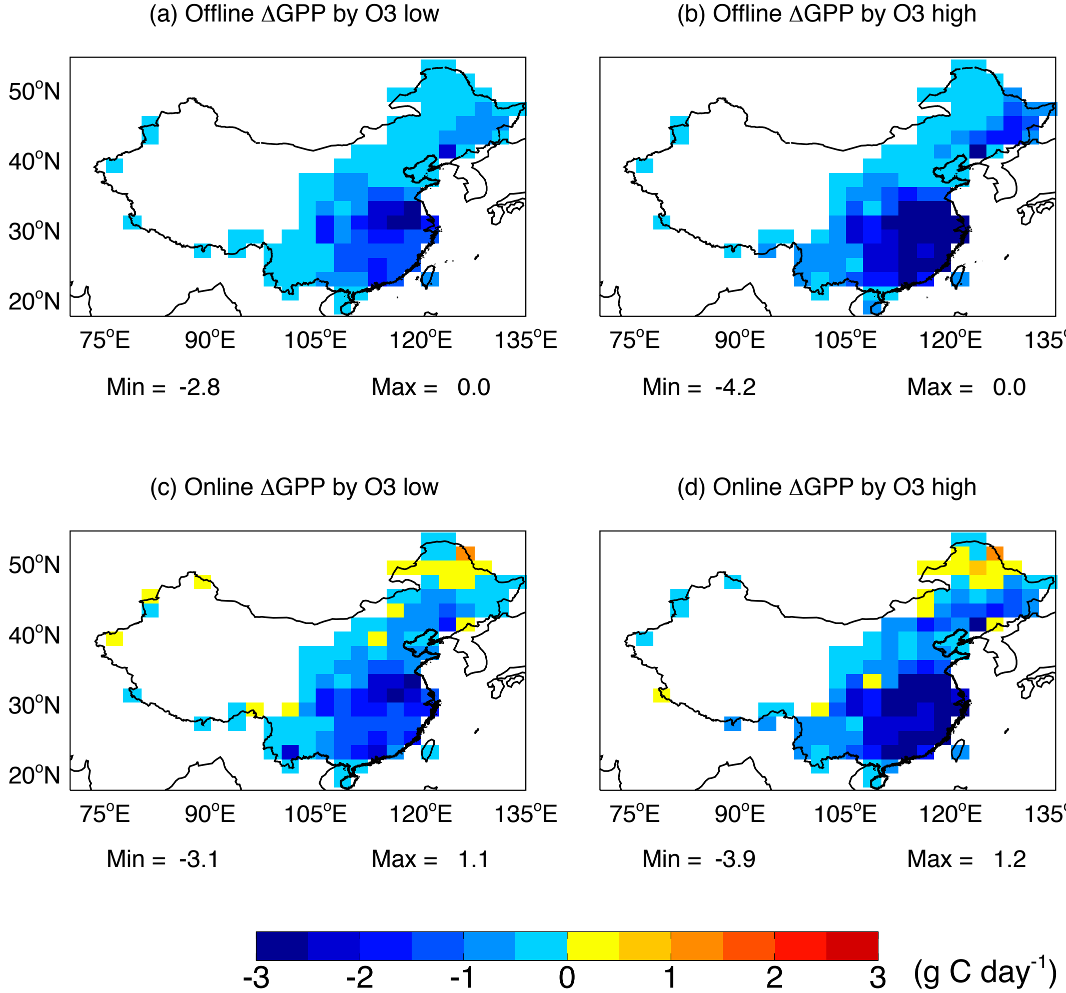
*Page 13, line 395. There doesn't seem to be any change in soil water in the North China Plain (fig S8f) in the same region where NPP decreases in fig S9d.*




→ The results shown in Fig. S9f (original Fig. S8f) are based on last 15-year simulations (years 6-20). However, results of offline simulations are based on 10-year meteorology (years 6-15). We show the changes in soil moisture for years 6-15 in the above. We can see that the pattern of soil water deficit in the above figure matches NPP changes in Fig. S10d (original Fig. S9d). The changes of soil water in North China Plain are not statistically significant in both the 15-year and 10-year simulations. The lack of significance may cause the inconsistency of NPP changes, i.e., the pattern of Fig. S10d, between the 10-year and 15-year simulations. However, the main conclusion that soil moisture plays the domain role in the NPP responses remains correct because NPP changes by temperature and radiation are far smaller than that by soil moisture. In the text, we added the following statement to remind readers about the discrepancies between online and offline simulations: "Uncertainties due to interannual climate variability in the model are calculated using different time periods for the online (15 years, Table 2) and offline (10 years, Table S3) runs." (Lines 311-313)

*Page 14, line 404. Is the agreement between the offline and online O3 inhibition true as a geographical pattern as well as the China total?*

→ Yes. We show both the online and offline damage below. Although we can see some differences at the individual grid cell level, the spatial patterns are quite similar between the online and offline runs. We do not present the figure in the paper because it is already busy with 10 Figures and supporting information. Instead, we state: "Application of ModelE2-YIBs that allows for these feedbacks gives an $O_3$-induced damage to annual GPP of 10.7%, a similar level to the damage computed in YIBs offline. The spatial pattern of the online $O_3$ inhibition also resembles that of offline damages (not shown)." (Lines 399-402)




*Page 14, line 406-407. Explain that the range quoted is for no AIE compared to AIE. Uncertainties should also be quoted and include the range between high and low sensitivity.*

→ We clarified as follows: "In the year 2010, the combined effects of $O_3$ and aerosols (Table 3) decrease total NPP in China by 0.39 (without AIE) to 0.80 Pg C yr$^{-1}$ (with AIE), …" (Lines 468-470)

We do not add uncertainties of $O_3$ sensitivity here to avoid redundancy and repetition. Instead, we include those uncertainties in a subsequent sentence: "Independently, $O_3$ reduces NPP by 0.59 Pg C yr$^{-1}$, with a large range from 0.43 Pg C yr$^{-1}$ for low sensitivity to 0.76 Pg C yr$^{-1}$ for high sensitivity (Table 3)." (Lines 476-478)

*Page 14, line 427. What is the change in methane in 2030?*

→ In the Methods section, we now describe the changes in $CH_4$ concentrations: "Well-mixed GHG concentrations are also adopted from the RCP8.5 scenario, with $CO_2$ changes from 390 ppm in 2010 to 449 ppm in 2030, and $CH_4$ changes from 1.779 ppm to 2.132 ppm." (Lines 264-266)

We agree that the increased global [$CH_4$] also contributes to higher [$O_3$]. We clarified the sentence as follows: "In contrast, the enhancement of [$O_3$] results from the increased $NO_x$ emissions, higher level of background $CH_4$ (~20%), and higher air temperature in the warmer 2030 climate." (Lines 491-493)

*Page 15, lines 434-436. It would be useful to be told the change in g_s between 2010 and 2030.*

→ We include the g_s change in the text: "Despite …, which contributes to $g_s$ inhibition of 4% on the country level, …". (Lines 500-501)

*Page 15, line 436. Need to include the high-low sensitivity range here.*

→ We clarified as follows: "…the future $O_3$-induced NPP damage in 2030 degrades to 14% or 0.67 Pg C $yr^{-1}$ (Table 3), with a range from 0.43 Pg C $yr^{-1}$ (low $O_3$ sensitivity) to 0.90 Pg C $yr^{-1}$ (high $O_3$ sensitivity)." (Lines 501-503)

*Page 15, line 441. Need to include the high-low sensitivity range here.*

→ We clarified as follows: "The model projects much lower damage to NPP of only 0.12 Pg C $yr^{-1}$, with a range from 0.06 Pg C $yr^{-1}$ (low O3 sensitivity) to 0.20 Pg C $yr^{-1}$ (high O3 sensitivity), mainly due to the 40% reduction in [O3] (Fig. 10c). Including both aerosol direct and indirect effects, O3 and aerosols together inhibit future NPP by 0.28 Pg C $yr^{-1}$, ranging from 0.12 Pg C $yr^{-1}$ with low O3 sensitivity to 0.43 Pg C $yr^{-1}$ with high O3 sensitivity." (Lines 507-512).

*Figure 3. The right hand plot needs a legend to explain the colours.*

→ A legend was added as suggested.

*Figures 4, 6, 8, 9, 10, S5, S7, S9: The south east China box should be shown in every panel.*

→ We prefer to show the box domain only in panel (a) of the figures to be concise and for improved clarity.

*Figure 4. The key for blue and black dots should be provided within the graphs.*

→ We tried to add the legend for blue and black dots on the scatter plot but found it difficult to place it because of the space limit. It is also inappropriate to place the legend outside the scatter plot because there are many coloured points in the middle panels (b and e). As a result, we do not make any changes to this Figure.

*Figure 5. The key for colours should be provided in the graphs. It would be useful to provide the letter keys within table S1. A different key may be better as there a several authors starting with "Z".*

→ A legend for symbol colours has been added for Figure 5. We added a new column to indicate the abbreviations of references shown in Figure 5. Different authors with the same initials have been differentiated with additional characters. For example, 'Fo' represents Foot et al. (1996).

*Figure 8. Clarify here and/or in the text that the PAR changes include the DRF.*

→ We have added the following sentence in the figure caption: "The NPP responses to PAR include the DRF effects."

*Figure S3. The key for colours should be provided in the graphs.*

→ A legend key has been added for Figure S3 as suggested.

*Figure S5. The colour scale for percentages should use the red colours for all the positive values, and blue only if there are negative ones, otherwise use the same colours as for the absolute values.*

→ We have changed the colour scale for percentages to the same ones as the absolute values in Figure S5.