

Reviewer #1

The authors are grateful to the editor and two reviewers for their time and energy in providing helpful comments that have improved the manuscript. In our revised paper, we added more explanations on methods and discussion, performed additional sensitivity tests, and moved original Figure S6 and S7 to the main text as new Figure 4 and 5 to improve the readability.

In this document, reviewers' comments have been addressed point by point. Referee comments are shown in black italics and author responses are shown in blue regular text. A manuscript with tracking changes is submitted separately.

*The manuscript by Hao Zhou and colleagues assesses the impacts of present-day aerosol loading on the scattering of shortwave radiation and its impacts on gross primary production in terrestrial ecosystems. It does so by using a suite of models: GEOS-Chem to simulate aerosol concentrations from multiple sources, CRM to compute the impacts of these aerosols on direct and diffuse shortwave radiation, and YIBs to simulate the impacts of direct/diffuse radiation on primary production.*

*Overall, this is an interesting study that attempts to break down the global-scale diffuse fertilization effect into contributions from different aerosol types and distinctions between anthropogenic and natural impacts. The topic is suited for publication in ACP, and assessments like the one presented here are helpful to shed light on the importance of the various aerosol-related impacts on radiation and productivity, and on the most important drivers of these impacts.*

→ We thank the reviewer for the positive evaluations.

*However, the description of the setup in the current manuscript is often presented in too concise a manner, which makes it hard to assess exactly how the impacts were computed and which other factors may play a role. Also, I have some questions about the setup that the authors might have thought of/addressed already in their setup, but that are not explained in the text. I list these major shortcomings below, followed by a list of minor comments that the authors could address when revising the manuscript.*

→ All the concerns raised by the reviewer has been carefully responded in this revised paper. Particularly, we added more explanations on our setup and moved some necessary figures from supplementary materials to main text for improving readability.

*Firstly, the study consequently expresses impacts of aerosols on PAR and GPP as increase or decrease, but it is not mentioned what the reference is for this. I trust that these are all expressed relative to the simulations without any aerosols, but I urge the authors to clarify this, and would recommend avoiding using “increase” or “decrease”, because these imply a trend in time and not an effect relative to a (hypothetical)*

reference case.

→ We understand the concerns from the reviewer on using “increase” and “decrease”, thus we further clarify this information “relative to simulations without aerosols” before expressing impacts of aerosols and use “enhance” and “reduce” to replace original “increase” and “decrease” in revised manuscript (e.g., lines 263-264).

*Also, the study makes a number of simplifications that are not spelled out in the text, but that I think should be explicitly mentioned and discussed. Most importantly, in the presentation of the results, the study treats the effects of different aerosols (anthropogenic/natural, BC/OC/Sulfate+Nitrate/Sea salt/dust) to be additive and independent, resulting in contributions to the radiation effects and GPP that nicely add up to 100% (l. 189ff, l. 316ff, Fig. 3, Fig. 4). However, because many non-linearities exist in the radiation responses to aerosols and the GPP responses to radiation, the sum of the individual effects will not be similar to the total effect, and individual effects are likely overestimated in the absence of other aerosols that can interfere with radiation. How has this been accounted for?*

→ In the revised paper, we clarified as follows:

For statistical methods used in the study:

“To evaluate the performance of models, we use statistical metrics including correlation coefficients (R) and normalized mean biases (NMB) defined as follows:

$$R = \frac{\sum_{i=1}^{i=n} (M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^{i=n} (M_i - \bar{M})^2 \times \sum_{i=1}^{i=n} (O_i - \bar{O})^2}} \quad (1)$$

$$NMB = \frac{\sum_{i=1}^{i=n} (M_i - O_i)}{\sum_{i=1}^{i=n} O_i} \quad (2)$$

where  $O_i$  and  $M_i$  are observed and modeled values, respectively.  $\bar{O}$  and  $\bar{M}$  are the averages of the observed and modeled values. In this study, R and NMB are used to evaluate the performance of models on the spatial scale, and Student t-test test is used to examine the significance of correlation coefficients and long-term trends.” (Lines 220-227)

For linear relationships between natural and anthropogenic aerosols:

“The GEOS-Chem runs GC\_ALL and GC\_NAT are driven with the same meteorology and emissions except that the former includes all sources of emissions while the latter excludes only anthropogenic emissions. Following the methods in Nascimento et al. (2021) and Ryu et al. (2013), we use the differences between GC\_ALL and GC\_NAT to represent aerosol concentrations contributed by anthropogenic sources. In this practice, the sums of natural and anthropogenic aerosol concentrations are equal to the total aerosol concentrations without non-linear effects.” (Lines 185-191)

For non-linear influences of varied aerosol species:

“It should be noted that such setup cannot resolve the interactive responses among

aerosol species, because the sum of individual aerosol effects are not necessarily equal to the net impact of all aerosols. The magnitude of these non-linear effects will be evaluated accordingly.” (Lines 202-205)

In the discussion section, we quantify the non-linear effects as follows:

“Fourth, we ignored the interactive effects among different aerosol species. Although we isolated the impacts of individual aerosol species on global GPP, their non-linear influences still exist in our simulations. For the radiative responses to aerosol species, we found that total aerosols enhance diffuse PAR by  $1.26 \text{ W m}^{-2}$  (Figure 2) and reduce direct PAR by  $2.78 \text{ W m}^{-2}$  (Figure S4). However, the sum of individual aerosol effects causes a net enhancement of  $1.35 \text{ W m}^{-2}$  in diffuse PAR (Figure S5) and a reduction of  $2.9 \text{ W m}^{-2}$  in direct PAR (Figure S6), both of which are slightly higher than the effects of all aerosols. Similarly, aerosols enhance global GPP by  $0.95 \text{ Pg C yr}^{-1}$  (Figure 3) but the sum of individual aerosol species enhance global GPP by  $1.21 \text{ Pg C yr}^{-1}$  (Figure 5). Such non-linearity is caused by the complicated responses of individual aerosol species, which can offset each other when they are put together. To facilitate the comparisons, we explore both the absolute (Figures 6c and 6d) and actual (Figures 6e and 6f) contributions of individual aerosol species to global GPP.” (Lines 503-515)

*Similarly, the YIBs setup for assessing the clear sky impacts on GPP should be explained in detail. Can YIBs be run with clear sky radiation only? And if this is done simply by assuming year-round clear sky impacts, how do you account for the changes in the vegetation state (e.g. change in LAI) that are simulated as a result of the additional growth? Please explain how clear-sky effects have been assessed in YIBs. Also, other forcing than meteorological should be described, notably the role of CO<sub>2</sub>. If observed CO<sub>2</sub> concentrations have been used, this might explain (part of) the trend in GPP displayed in Fig. 5ab.*

→ Yes, the YIBs model can be run with clear-sky radiation alone simulated by CRM model. In this study, all forcings except radiation are adopted from observations or climate reanalyses. For the original Fig. 5 (now Fig. 7), the results shown are the differences between simulations with and without aerosols. As a result, the CO<sub>2</sub> fertilization effects have been excluded and the trend of GPP shown is attributed to the differences of aerosol-induced radiation, instead of CO<sub>2</sub> concentrations.

We clarified as follows:

“Land cover product from MODIS is used as vegetation coverage for YIBs model (Yue et al., 2021) and observed CO<sub>2</sub> concentrations from Mauna Loa are also used (Yue et al., 2015).” (Lines 178-180)

“For YIBs runs, other forcings (e.g., CO<sub>2</sub> concentrations and climate meteorology) except diffuse and direct PAR are kept the same in all runs, so as to exclude their impacts on global GPP.” (Lines 207-209)

*Lastly, the manuscript is nicely written, but it heavily relies on material published in the supplementary information. I think that it is generally a good idea to provide*

*supplementary material, but in the current manuscript, it is often “need to have” (rather than “nice to have”) material that is in there. This results in a manuscript with a very large number of references to the supplementary information. I would recommend considering whether the text can be understood without access to the figures in the supplementary information. For those figures where this is not the case, I suggest promoting the figures into the main text.*

→ Thank you for your helpful suggestions. We moved the original Figures S6 and S7 to the main text as new Figures 4 and 5, which had been cited frequently.

*Given these shortcomings, I cannot recommend the current study for publication in ACP, but I would like to encourage the authors to address these in a revised version of the manuscript.*

→ In the revised paper, we have made necessary improvements as suggested by the reviewer.

*Minor remarks:*

*L. 28: consider replacing “ratio” by “fraction”*

→ Corrected as suggested.

*L. 42: I do not agree with this explanation of the DFE! Enhanced GPP is not so much an effect of changes of the LUE of shaded leaves, but rather an increase of the fraction of shaded leaves (that have a higher LUE than sunlit leaves because of the lower PAR levels), hence increasing the total canopy LUE.*

→ We had revised our explanation on DFE as followed:

“The cause for such difference is that diffuse light can penetrate into the deep canopy and enhance photosynthesis of more shaded leaves with higher light use efficiency ( $LUE = GPP/PAR$ , gross primary production per photosynthetically active radiation) (Roderick et al., 2001; Gu et al., 2003; Rap et al., 2015).” (Lines 43-46)

*L. 119: Please explain here why you do two sets of simulations (one with CEDS, and one with EDGAR) – it is not clear why you need two alternatives, and how you will treat them in the manuscript.*

→ We used two different emission inventories to assess the modeling uncertainties due to emission inventories. We have clarified as follows:

“Second, the uncertainties of emission inventory may influence the conclusions. In this study, CEDS emission inventory is used for anthropogenic emissions. Here, we used

another emission database (EDGAR) to assess the uncertainties of DFE from anthropogenic aerosols. The new simulations showed that anthropogenic aerosols increased global GPP by 0.31 Pg C yr<sup>-1</sup> (Figures S13-S14), lower than the value of 0.39 Pg C yr<sup>-1</sup> predicted with CEDS inventory (Figure 3). The spatial pattern of the percentage contributions remains similar for the two inventories, both of which show dominant impacts by anthropogenic aerosols over Eastern China, India, Europe and North America. For DFE of aerosol species, anthropogenic sulfate and nitrate aerosols still dominate global aerosol DFE up to 28.2 % and natural OC aerosols contribute 18.2% to aerosol DFE (Figure S15), which is similar to that from CEDS.” (Lines 477-487)

*Eqs. (1) and (2): The computation of sunlit fraction from Beer-Lambert’s law implies that not all light will be absorbed by a canopy, and in particular with low L, F<sub>sunlit</sub>+F<sub>shaded</sub> could be considerably lower than 1. Also, please clarify whether the described treatment of sunlit and shaded leaves is standard in YIBs or whether it was altered for this study.*

➔ Thank you for your question. In this study, we applied the canopy radiative transfer scheme proposed by Spitters, et al. (1986) to distinguish the responses of shaded and sunlit leaves to diffuse and direct radiation. In this scheme, the fraction of shading (F<sub>shaded</sub>) and sunlit (F<sub>sunlit</sub>) leaves is changing dependent on both solar zenith and leaf area index (Equation 2). The sum of F<sub>shaded</sub> and F<sub>sunlit</sub> is always 1, but the amount of radiation, both diffuse and direct components, is dampened with the penetration of light into the deep canopy.

Yes, not all light will be absorbed by the canopy due to the extinction processes. In our parameterization, such incompleteness is reflected in photosynthesis of shaded (A<sub>shaded</sub>) and sunlit (A<sub>sunlit</sub>), which are changing all the time to reflect the impacts of light extinction. Figure 1 shows that this canopy radiative transfer scheme reasonably captures the different responses of GPP to direct and diffuse radiation.

We clarified as follows: “Compared with global in situ measurements, this canopy radiative transfer scheme reasonably captures the different responses of GPP to direct and diffuse radiation (Yue and Unger, 2018;Zhou et al., 2021a). For this study, we use the original scheme without modifications.” (Lines 166-167).

*L. 208ff: Please provide more information in the main text about the observations, so that it can be understood without the supplementary information at hand. Specifically, please explain how the correlation coefficients and NMBs were determined (L. 210, 218, 219, etc): Is it based on spatial variations or temporal, and if the latter is considered, at which timescale (monthly?) is the temporal variation assessed? What statistical test was used to determine R values?*

➔ We add more details into methods as follows:

“To evaluate the performance of models, we use statistical metrics including correlation coefficients (R) and normalized mean biases (NMB) defined as follows:

$$R = \frac{\sum_{i=1}^{i=n} (M_i - \bar{M})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^{i=n} (M_i - \bar{M})^2 \times \sum_{i=1}^{i=n} (O_i - \bar{O})^2}} \quad (1)$$

$$NMB = \frac{\sum_{i=1}^{i=n} (M_i - O_i)}{\sum_{i=1}^{i=n} O_i} \quad (2)$$

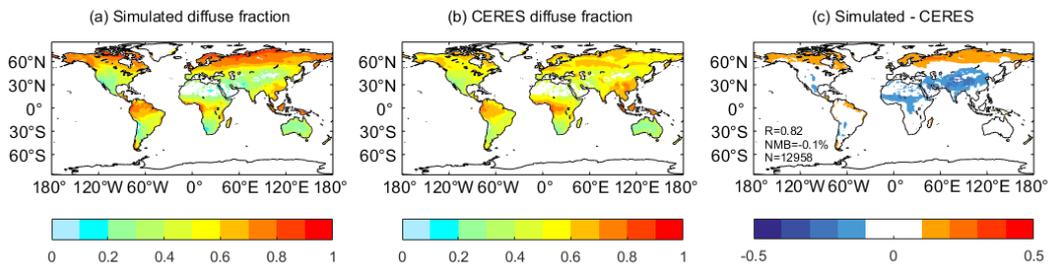
where  $O_i$  and  $M_i$  are observed and modeled values, respectively.  $\bar{O}$  and  $\bar{M}$  are the averages of the observed and modeled values. In this study, R and NMB are used to evaluate the performance of models on the spatial scale, and Student t-test test is used to examine the significance of correlation coefficients and long-term trends.” (Lines 220-227)

All the evaluations in this study are based on the spatial variations. We clarified in the Figure captions that all the results are shown on the annual mean basis. In our previous studies, we evaluated the temporal variations of simulated GPP (e.g., Tian, et al. (2021) and Yue, et al. (2020)).

*L. 216: It is no surprise to obtain high R values when comparing simulated and observed magnitudes of SW radiation when considering that there is a clear latitudinal dependency that is present already in the top-of-atmosphere estimates. In order to assess the impacts of the radiative transfer scheme specifically, the authors may want to choose a more specific metric to evaluate its performance, e.g. the fraction of diffuse radiation.*

➔ Thank you for your suggestion. We added the validation of simulated diffuse fraction as follows (also see Figure S3):

“Although the CRM model presents high R and low NMB under both sky conditions, evaluations still show that modeled shortwave radiation is higher than observations. Such overestimation may be related to the underestimation of simulated AOD (Figure S1), which leads to more shortwave radiation reaching the surface. We further evaluate the simulated diffuse fraction (DF) with satellite observations (Figure S3). Simulations reproduce observed spatial pattern with high R of 0.82 and low NMB of -0.1% on the global scale, but overestimate regional DF over high latitudes and underestimate DF over Asia.” (Lines 242-249)



**Figure S3** Evaluations of simulated diffuse fraction (DF) by CRM model. Results shown are the annual DF from (a) simulations, (b) observations and (c) their differences over land vegetated grids during 2001-2014. Simulations are performed using the Column Radiation Model (CRM), which is driven with hourly  $1^{\circ}\times 1^{\circ}$  meteorological forcings from MERRA-2 and cloud profiles from the SYN1deg product of CERES. The R, NMB and N are shown in (c).

*Fig. 2: Please consider using the same colour scale for all panels in the figure.*

→ Corrected as suggested.

*L. 318: Replace “grids” with “of the grid cells”*

→ Corrected as suggested.

*L. 331: Please mention the statistical test used to determine significance of the trend. Also, are numbers used for DFE here expressed as changes in GPP?*

→ Yes, changes of DFE are equal to GPP changes by aerosols in this study, and we clarify statistical test in this study as followed:

“Student t-test test is used to examine the significance of correlation coefficients and long-term trends.” (Lines 226-227)

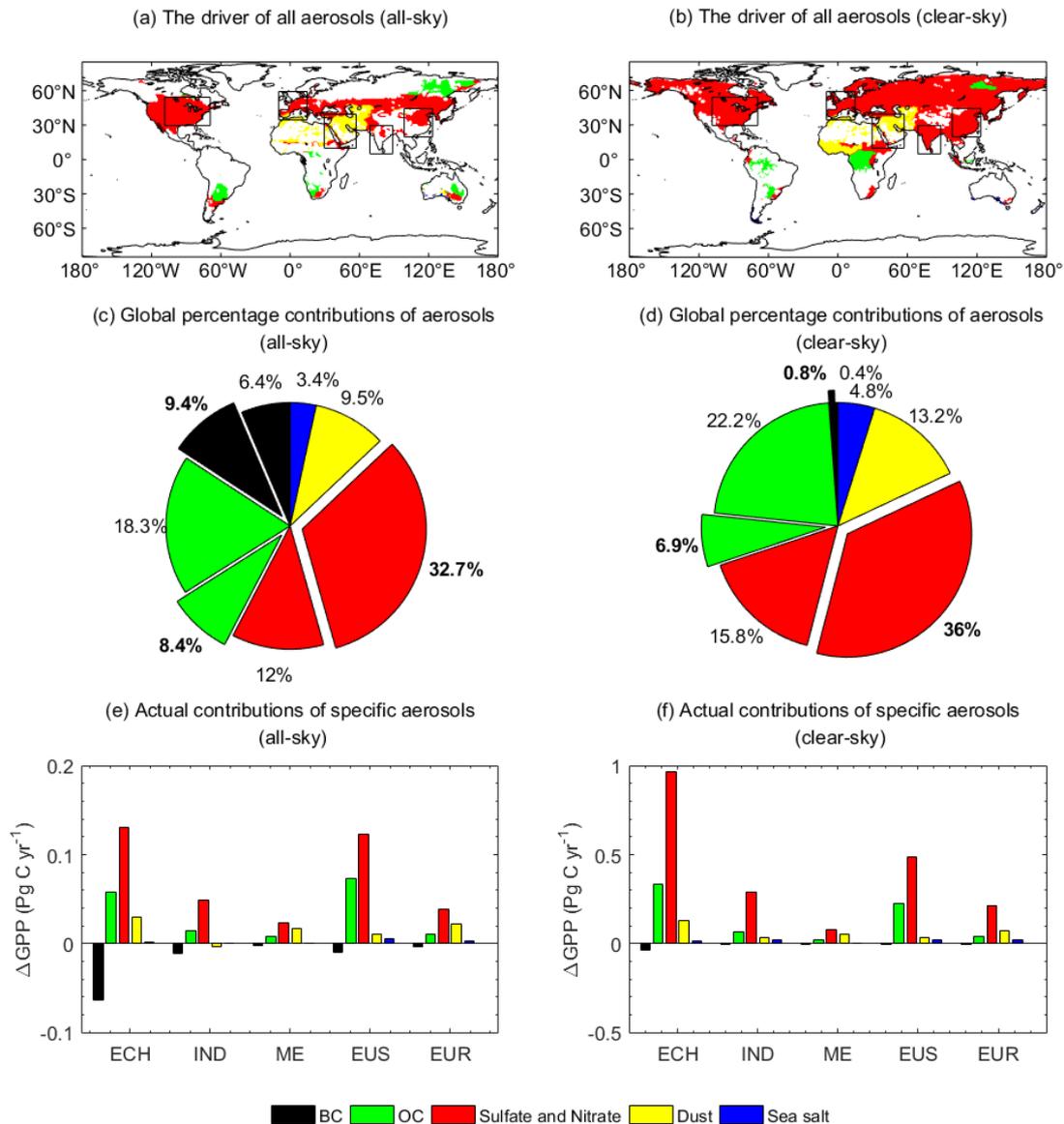
*L. 446-448: Consider replacing “cloud” by “clouds”*

→ Corrected as suggested.

*Fig. 4cd: I am not so fond of the computation of the contribution of individual drivers based on absolute amounts – I think it is important to stress (also graphically in the figure) that BC has a negative, and all other aerosols a positive impact on GPP. See also my comments above about the computation of these contributions.*

→ Thank you for your suggestion. In the revised paper, we used both the absolute (to indicate the magnitude) and actual (to indicate the signs) contributions of individual aerosol species (see the updated Figure 6 shown below).

To emphasize the contributions of BC aerosols to total aerosol DFE, we still select percentage contributions of absolute amounts as a metric of DFE from individual aerosol species (Figures 6c and 6d). Meanwhile, we replace the original (e) and (f) with actual DFE to indicate the negative effects of BC aerosols.



**Figure 6** (a, b) Dominant aerosol species contributing to the simulated changes in annual GPP, (c, d) percentage contributions of aerosol species to global GPP, and (e, f) actual DFE of aerosol species in specific regions at (a, c, e) all skies and (b, d, f) clear skies. The contributions in (c) and (d) are calculated as the ratios of absolute DFE, as BC aerosols induce negative DFE. The normal (bold) fonts in (c) and (d) represent aerosol species from natural (anthropogenic) sources. Regions with relatively high percentage changes in GPP (>1% for all-sky and >5% for clear-sky) by aerosols are shown in (a) and (b). The regions include eastern China (ECH), India (INA), Middle East (ME), eastern U.S. (EUS), and Europe (EUR), which are marked as black boxes in (a) and (b). The black, green, red, yellow, and blue represent the effects of BC, OC, sulfate and nitrate, dust, and sea salt aerosols, respectively.

**Reference:**

Spitters, C.J.T., Toussaint, H., Goudriaan, J., 1986. Separating the diffuse and direct component of global radiation and its implications for modeling canopy photosynthesis Part 1. components of incoming radiation. *Agricultural and Forest Meteorology* 38, 217-229.

Tian, C., Yue, X., Zhou, H., Lei, Y., Ma, Y., Cao, Y., 2021. Projections of changes in ecosystem productivity under 1.5 °C and 2 °C global warming. *Global and Planetary Change* 205, 103588.

Yue, X., Liao, H., Wang, H., Zhang, T., Unger, N., Sitch, S., Feng, Z., Yang, J., 2020. Pathway dependence of ecosystem responses in China to 1.5 degrees C global warming. *Atmos Chem Phys* 20, 2353-2366.