Reviewer #2

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.

General comments:

This study used the GEOS-Chem chemistry transport model, combined with the CRM radiation model and the Yale Interactive terrestrial Biosphere (YIBs) model to quantify the impact of 2001-2014 global aerosol distributions on gross primary productivity via the diffuse radiation fertilization effect. The paper addresses an interesting topic and could bring an important contribution to existing literature in this area. However, in my opinion it still requires some important revisions before it can be published.

➔We thank the reviewer for the positive evaluations.

Major comments:

Quantifying the uncertainty of the estimates presented. While Section 4.2 does acknowledge the limitations and uncertainties of the study, more should be done to quantify the effect of these uncertainties on the calculated changes in radiation and GPP. For example, how does the underestimation of simulated AOD compared to MODIS (e.g. Fig. S1) affect the calculated changes in GPP (i.e. error bars on the 0.95 Pg C yr-1 estimate for GPP increase caused by aerosol diffuse radiation fertilization)? And, in particular, what is the effect of the reported substantial bias in some key regions such as the Amazon, central Africa and boreal Asia (lines 413-415)?

➔Thank you for the suggestion. We added three simulations including 1.5, 2 and 3 times of original aerosol concentrations to explore the uncertainties of aerosol DFE (also see Figure S12).

"To explore the effects of such underestimation on global aerosol DFE, we performed three additional simulations with 1.5, 2 and 3 times of original aerosol concentrations. Predicted aerosol DFE in these three simulations are respectively, 1.13 Pg C yr^{-1}, 1.18 Pg C yr^{-1} and 0.97 Pg C yr^{-1} (Figure S12), similar to the estimate of 0.95 Pg C yr^{-1} (Figure 3a) with original aerosol concentrations. Regionally, aerosols reduce GPP up to -3% over Amazon, Center Africa, India, eastern China and Indonesia under double or tripled aerosols conditions, which are related to negative effects from high cloud amount (Figure S11) or aerosol loading (Figure S1).” (Lines 468-476)

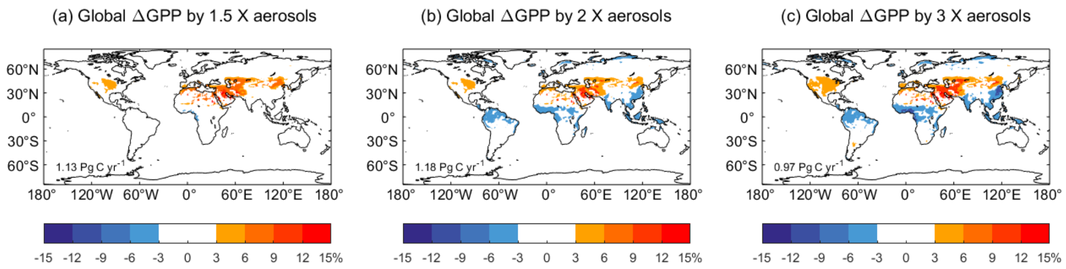

Figure S12 The same as Figure 3a but with 1.5, 2, and 3 times of aerosol concentrations.

Linearity of all simulated effects. Figure 1 indicates a highly non-linear GPP to PAR response. Nevertheless, all results presented show the opposite, with linear responses for both the PAR to aerosol loading response (e.g. Fig. 2, Fig. S3), and for the GPP to aerosol loading response (e.g. Fig. 3, Fig S6). This should be discussed and clearly justified.

➔In the revised paper, we added more explanations on linear or non-linear effects in our simulations.

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 sum 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 W m^{-2} (Figure 2) and reduce direct PAR by 2.78 W m^{-2} (Figure S4). However, the sum of individual aerosol effects causes a net enhancement of 1.35 W m^{-2} in diffuse PAR (Figure S5) and a reduction of 2.9 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 Pg C yr^{-1} (Figure 3) but the sum of individual aerosol species enhance global GPP by 1.21 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)

While the structure of the paper is relatively clear, I found it quite hard to follow the argument in some cases. I suggest a careful rewrite of some of the results paragraphs, with a more clear highlight of the main results presented. Also, to improve readability of the paper, some figures from the supplementary material might be better suited in the main manuscript.

➔ 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. We also added a summary statement at the beginning of most paragraphs to increase the readability of the paper.

Specific comments:

How do the aerosol induced changes in PAR compare with other published results? How realistic are the results presented in Figures 2 and 3? For example the large areas with virtually zero aerosol effect on diffuse PAR (e.g. South America, Australia, large parts of Europe, Northern Asia).

➔ The large areas with blank changes are actually non-zero. It is white because the regional perturbations in diffuse radiation are smaller than 3 W m^{-2}. In the revised paper, we added the following discussion:

"Regionally, aerosols cause large enhancement of diffuse PAR (>3 W m^{-2}) over southern U.S., Australia, Europe, and northern Asia under clear sky conditions (Figure 2d). However, these enhancements of diffuse PAR are largely dampened under all sky conditions (Figure 2a). Similar changes in diffuse radiation by aerosols are predicted by Chen and Zhuang (2014) and Rap et al. (2018), though the former study yielded much larger changes in radiation and the latter examined only biogenic aerosols. The cause of smaller PAR changes under all sky conditions is that cloud tends to weaken aerosol radiative forcing by amplifying absorption and diminishing scattering (Paulot et al., 2018)." (Lines 269-275)

More details should be provided for the evaluation of the CRM radiative transfer model. What causes the differences in simulated and observed clear-sky and all-sky SW fluxes presented in Figure S2 (i.e. a vs. b, d vs. e, a-d vs b-e), considering that both the model clouds (lines 147-149) and the SW fluxes used for validation (lines 203-205) are based on CERES SYN1deg observations?

➔ In the revised paper, we discussed the performance of CRM model, and added the validations of simulated diffuse fraction (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)

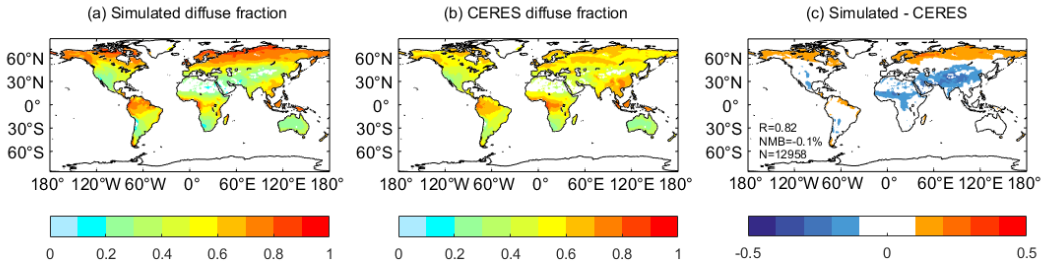

Figure S3 Evaluations of simulated diffuse fraction (DF) by CRM model. Results shown are the annual DF from (a) simulations, (b) observations and (c) their differences over land vegetated grids during 2001-2014. Simulations are performed using the Column Radiation Model (CRM), which is driven with hourly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

Should include a more thorough discussion of the implications and meaning of the various estimates presented under clear sky and under all sky conditions, rather than simply listing these values.

Why is the 2003 difference between simulated all-sky GPP changes from natural and anthropogenic aerosols the smallest (Fig 5a), while the same difference under clear sky conditions seem to be the largest across all period (Fig 5b)?

➔We have included the discussion about the cloud effects on aerosol DFE and its contributions to global trends in GPP changes:

“In our simulations, aerosols increase global GPP by 8.91 Pg C yr^{-1} under clear sky conditions but only 0.95 Pg C yr^{-1} under all sky conditions. Similarly, Cohan et al. (2002) and Yue and Unger (2017) found aerosol DFE was limited at cloudy skies. Cloud can mask aerosol DFE by modifying both the quantity and quality of aerosol radiative perturbations (Yu et al., 2006). First, cloud weakens the impacts of aerosols on both direct and diffuse radiation (Figures 2 and S4) by reducing the total sunlight available for the extinction by aerosols (Kinne, 2019). Therefore, the smaller changes in diffuse PAR by aerosols under all sky conditions (Figure 2) result in lower DFE than that under clear sky conditions. Second, cloud significantly reduces direct radiation and limits the potential of increasing GPP by diffuse radiation. Observations have shown an optimal diffuse fraction of 0.4-0.6 to enhance GPP for most plant types (Zhou et al., 2021c). A further increase of diffuse fraction above the optimal range will dampen GPP due to the reduced photosynthesis of sunlit leaves. Appearance of cloud has provided an environment with high diffuse fraction that aerosols may have limited benefits or even negative effects for GPP (Yue and Unger, 2017). Such relationship also explains why the decreasing trend of global cloud amount contributes to an increased aerosol DFE (Figure 7a)." (Lines 403-419)

We added the following discussion to explain why the year 2003 is a turning points between DFE from natural and anthropogenic aerosols.
"The differences between natural and anthropogenic aerosol DFE are inconsistent at varied sky conditions (Figure 7). For the year 2003, ΔGPP by natural aerosols is very close to that by anthropogenic aerosols under all-sky conditions (Figure 7a). However, the same year sees large differences of ΔGPP between different sources of aerosols at clear-sky conditions (Figure 7b). Analyses show that increased cloud amount weakens aerosol DFE especially over central Africa and boreal Asia with high loading of natural aerosols before 2003 (Figure S11a), but decreased cloud amount enhances natural aerosol DFE over Amazon, central Africa, and boreal Asia after 2003 (Figure S11b). These opposite trends of cloud over regions with high loading of natural aerosols lead to a turning point for natural aerosol DFE in 2003 under all-sky conditions." (Lines 389-398)

To better put these estimates into perspective, it would be very useful to also provide an estimate of the magnitude of other (non-included) aerosol effects, in particular the aerosol-induced changes in temperature? This could be done by performing an additional YIBs simulation driven by aerosol induced temperature changes estimated using existing aerosol transient climate sensitivity values.

➔ Thank you for your suggestions. The aerosol-induced temperature changes are dependent on radiative properties of aerosol species, surface albedos, and the feedback of climate system. As a result, it is not a linear relationship to aerosol optical depth or radiative forcing. In addition, the temperature changes by aerosols are very uncertain among different models. It is not reasonable to apply specific climate sensitivity values to derive the aerosol-induced temperature changes.

In the revised paper, we compared aerosol DFE with other aerosol effects simulated in previous studies:
"In our previous studies, we explored the direct aerosol radiative effects on NPP in

China through changes in radiation, temperature and soil moisture, and found that aerosol DFE enhances regional NPP by 0.09 Pg C yr^{-1} which accounts for ~50% of the total aerosol effects (Yue et al., 2017b). Similarly, Zhang et al. (2021) explored the impacts of anthropogenic aerosols on global carbon sink during 1850-2014, and found that aerosol DFE accounts for 78% of the total aerosol effects on carbon uptake, which is much higher than the effects caused by temperature and precipitation changes." (Lines 522-528)

***Technical corrections**:*

Line 180: "source" should be "sources".

➔Corrected as suggested.

Line 211: "normalize" should be "normalized"

➔Corrected as suggested.

In various places (e.g. lines 24-25) "at clear skies" or "at all skies" does not read well and could be changed to e.g. "under clear sky conditions".

➔Corrected as suggested.

Clearly state how the standard deviation illustrated in Figure 5 was calculated.

➔we added the explanation of standard deviation in caption of Figure 7 (original Figure 5): "The hollow circles and shadings in (a) and (b) represent annual mean and standard deviation of aerosol-induced GPP changes from all months in each year."