Reply to Anonymous Reviewer #2:

We appreciate the reviewer's comments on the manuscript. All comments are highly valuable and helpful for us to improve our manuscript. We have studied them carefully and have addressed them in the revised manuscript. Below are point-by point responses to the reviewer's comments.

Comments from the reviewer:

1. The authors have done a good job in analyzing the aerosol distribution in both spatial and temporal domain. As a step forward, it would be interesting to examine the spatiotemporal variations of the absorbing capability of the aerosols from the wildfires in Australia. Since the AERONET has single scattering albedo (SSA) as a product, it would be a low-hanging fruit to analyze it. In particular, how well the BC and OC from MERRA2 are correlated with the observed SSA is of great interest.

We appreciate these valuable suggestions. Following these suggestions, we have performed the analysis of monthly averaged single scattering albedo (SSA) at the nine AERONET sites over Australia. The BC and OC from MERRA2 are correlated well with SSA from AERONET. For example, we found that SSA values at Lake Argyle were lower (< 0.90 at 440 nm wavelength) during September-November, suggesting the relative dominance of absorbing aerosols such as dust and black carbon. Furthermore, the northwestern Australia had always been the relatively high carbonaceous (BC+OC) AOD region during BB period, which is consistent with the assimilation from MERRA-2. In addition, we have added the discussions in our revised manuscript at several parts. For example,

Lines 251-260: "**During the BB period, fires cause a temporary reduction in vegetation cover, which can increase biomass burning emissions which are primarily fine aerosol particles. SSA also showed decreasing trends with increase in wavelengths in most months at Jabiru and Lake Argyle in northwestern Australia, especially during the BB period (Figs. 6a, 6b), which showed stronger absorption in the near-infrared bands. Fires also accelerate soil erosion by winds and promote dust emissions (Ravi et al.,2012). The coarse particles such as dust in northern Australia could have been entrained into the biomass burning plume from local soil and also been transported from central Australian deserts (Winton et al., 2016; Yang et al., 2020b). SSA values at Lake Argyle were lower (< 0.90 at 440 nm wavelength) during September-November, suggesting the relative dominance of absorbing aerosols such as dust and black carbon. Furthermore, the sea salt aerosols from ocean would also contribute to the differences in volume size distributions and SSA among various sites, or between BB and non-BB periods.**".

Lines 267-273: "**SSA showed an ambiguous wavelength dependence (i.e., increasing or decreasing with wavelengths) at Learmonth due to the presence of aerosol mixture (Fig. 5c). However, the average SSA values were less than 0.90 at 440 nm wavelength during late spring and summer at Learmonth, showing**

absorbing properties of coarse particles, which was associated with the site's location in the North-Western dust pathway from the Australian interior deserts (e.g. the Gibson Desert and Great Victoria Desert). The average SSA values generally decreased with increasing spectral range at Lake Lefory possibly due to the anthropogenic emissions and biomass burnings (Yang et al., 2020) (Fig. 5d).”

Line 279-281: “**In eastern Australia, the coarse mode aerosols were dominant in almost all seasons at Lucinda (Fig. 5h). The average SSA values generally increased with increasing spectral range with low values (<0.95) at Lucinda (Fig. 6h).**”.

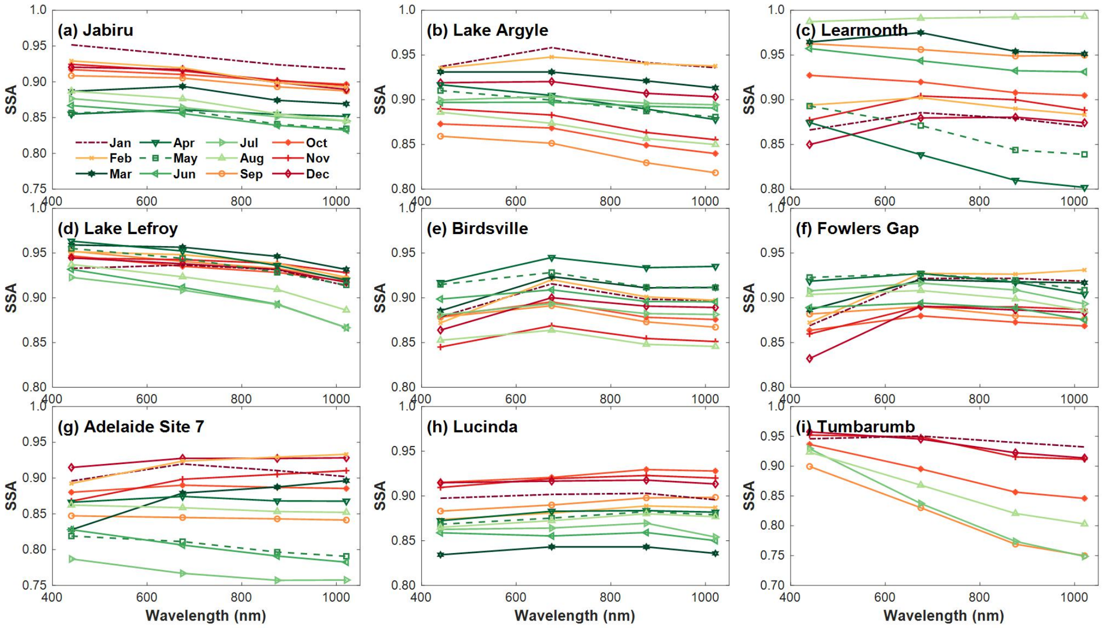

Figure 6. Monthly averaged single scattering albedo (SSA) at the nine AERONET sites over Australia. The warm-toned and cold-toned lines represent the aerosol size distributions in BB period and non-BB period, respectively. Note: Only SSA data from Tunbarumb is used due to the lack of SSA data at Canberra.

2. Please label out years in Figure 2.

Thank you for your suggestion. We have revised the Figure 2 as suggested.

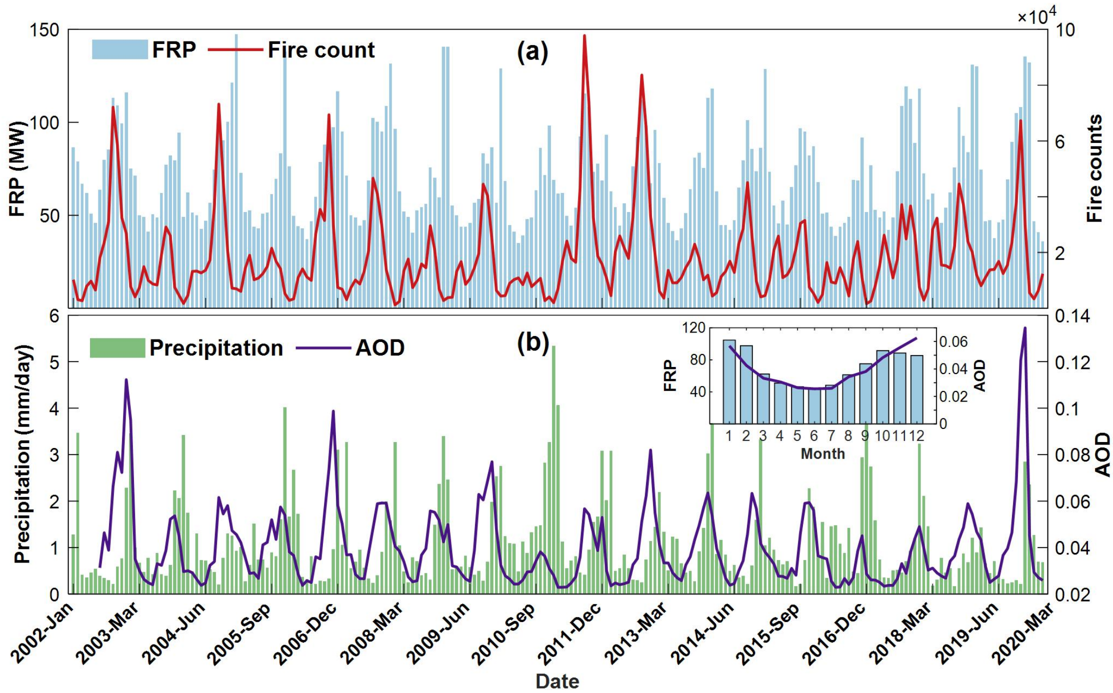

Figure 2. Time series of monthly averaged fire radiative power (FRP), fire count from MCD14ML (a), and total precipitation from ERA-5 and Aqua MODIS DB AOD (b) in Australia.

3. How are the aerosol volume size distributions measured by the AERONET? If they are from remote sensing retrievals, how reliable they are? Some references are needed here.

These are good questions and comments. The volume particle size distribution dV(r)/dlnr (μm^3 /μm^2) is retrieved for 22 logarithmically equidistant discrete points (ri) in the range of sizes 0.05 μm≤r≤15 μm. The Cimel sky radiance measurements in the almucantar plane at 440, 675, 870, and 1020 nm (nominal wavelengths) in conjunction with the direct sun measured AOD at these same wavelengths were used to retrieve column-integrated aerosol size distributions (dV(r)/dln(r) from 0.05 to 15 μm).

$$\frac{dV(r)}{dlnr} = V(r)\frac{dN(r)}{dlnr} = \frac{4}{3}\pi r^3 \frac{dN(r)}{dlnr}$$

We have added related references to describe the retrieval method and accuracy of aerosol volume size distributions from AERONET in the revised manuscript:

Lines 107-113: **"The retrievals of aerosol microphysical properties such as particle volume size distribution (dV(r)/dlnr) and single scattering albedo (SSA) are used in this study. The detailed retrieval algorithm can be found in Dubovik et al. (2000) and Dubovik et al. (2006), and hence are not reintroduced in this paper. The retrieval errors of dV(r)/dlnr did not exceed 10% in the maxima of the dV(r)/dlnr and may increase up to 35% for the minimum values of the dV(r)/dlnr in the intermediate particle size range (0.1 ≤ r ≤ 7 μm). The retrieval error of dV(r)/dlnr increased significantly for the edges of the particle size interval but did not significantly affect the derivation of the main features of the**

particle size distribution (Dubovik et al., 2002)."

Dubovik, O., Smirnov, A., Holben, B. N., King, M. D., Kaufman, Y. J., Eck, T. F., and Slutsker, I.: Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) Sun and sky radiance measurements, Journal of Geophysical Research: Atmospheres, 105, 9791-9806, 10.1029/2000jd900040, 2000.

Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman,Y. J., King, M. D., Tanré, D., and Slutsker, I.:Variability of Absorption and Optical Properties of Key Aerosol Types Observed in Worldwide Locations, J. Atmos. Sci., 59, 590–608,https://doi.org/10.1175/1520-0469(2002)059<0590:voaaop>2.0.co;2, 2002.

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B. N., Mishchenko, M., Yang, P., Eck, T. F., Volten, H., Muñoz, O., Veihelmann, B., van der Zande, W. J., Leon, J.-F., Sorokin, M., and Slutsker, I.: Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, Journal of Geophysical Research: Atmospheres, 111, https://doi.org/10.1029/2005JD006619, 2006.

4. About the comparison of AOD between MERRA2 and AERONET, I assume AERONET only report AOD in the non-cloudy days, while MERRA2 can calculate AOD anytime. Such a sampling issue needs to be mentioned.

We agree with the reviewer and have made corresponding changes in the revised manuscript, which are in Lines 164-166: "**In this study, the quality controlled and cloud screened AOD from AERONET was used to evaluate the performance of MERRA-2 AOD for only cloud-free condition, while MERRA-2 AOD provides the aerosol information in both cloud-free and cloudy conditions.**".

5. Fig. 8, please clarify at which level the mass concentrations are. Near surface?

We now have clarified the level of the mass concentrations which are surface mass concentrations, which are: "**Figure 8. Spatial distribution of carbonaceous, dust, sulfate, and sea salt mass concentrations near surface (a-h) and AOD (i-p) estimated by MERRA-2 during BB period and non-BB period over Australia.**".

6. MERRA2-Aero has black carbon and organic carbon separately. Does the term "carbonaceous" in the paper refers to the summation of those two species? Please clarify.

We appreciate this suggestion. We have clarified the data source of carbonaceous, which are in Lines 309-311: "**Considering the similar emission sources of organic carbon and black carbon aerosols, carbonaceous aerosol is used in this study to refer to the summation of organic carbon and black carbon from MERRA-2.**".

7. What is the source of dust detected at heights from 2 to 5 km in November 2019, January, and February 2020? From fire or from the desert in the west?

The reviewer proposed a good question. We simulated the backward trajectories at Tumbarumba, which is located at southeastern Australia, at 1000 m above ground level. The backward trajectories showed that the airflows were from the Lake Eyre Basin during the period September 2019- February 2020 (Fig.R1). In addition, Wagner et al. (2018) indicated that fire radiative energy released by the combustion of the vegetation leads to a significant increase in near-surface wind speed, atmospheric turbulence, and vortices. Moreover, the removal of vegetation during the burning process and the accompanied dehydration and modification of the soil could consequently enhance the dust mobilization and uplift potential, which finally influenced the concentration and the mean size of aerosol particles over the fire region. Therefore, the increase of the occurrence frequency of dust in November 2019, January 2020, and February 2020 is most likely the combination result of fire-induced dust emissions caused by the pyro-convection during extreme fire events and long range transport of dust from the Lake Eyre Basin. Of course, it is challenging for us to quantify their relative contribution. We added a description about this at Lines 392-401: "**Wagner et al. (2018) indicated that fire radiative energy released by the combustion of the vegetation leads to a significant increase in near-surface wind speed, atmospheric turbulence, and vortices. Moreover, the removal of vegetation during the burning process and the accompanied dehydration and modification of the soil could consequently enhance the dust mobilization and uplift potential, which finally influenced the concentration and mean size of aerosol particles over the fire region. McGowan and Clark. (2008) showed that dust from the Lake Eyre Basin can also potentially affect the southeastern Australia through the southeast dust transport corridor. Therefore, the increase in the occurrence frequency of dust explained the relatively high coarse mode aerosol volume concentrations at the sites in southeastern Australia (Fig.5), which was most likely a combination result of the fire-induced dust emissions caused by the pyro-convection during extreme fire events and long range transport of dust from the Lake Eyre Basin.**".

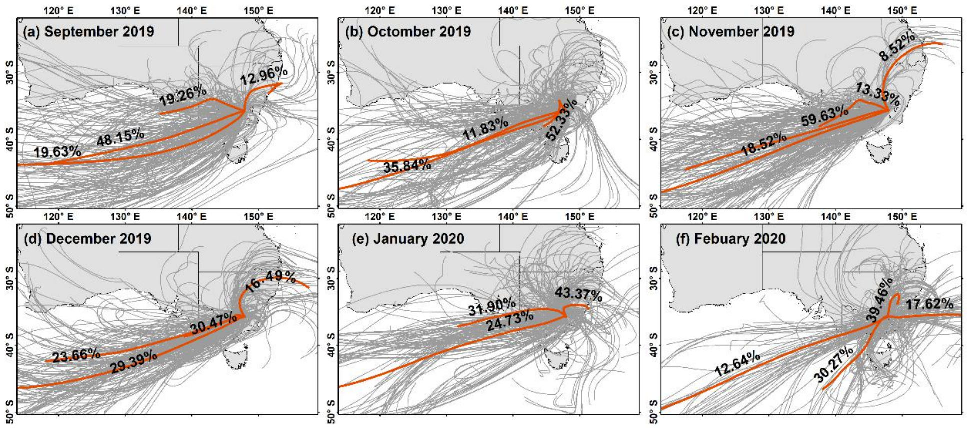

Fig.R1 The 72 h back trajectories ending at Tumbarumba at 1000 m above ground level. The gray lines represent the total back trajectories in each month. The orange lines represent the cluster back trajectories.