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. Abstract: Line 13-16: Please specify what time periods the trends are for. Line 22-26: Please specify which heights these descriptions refer to since you are talking about 0.5-5 km heights in the sentence that follows. Line 26-27: Which region does this sentence refer to? Line 27-28: The meaning of this sentence is very vague. Line 28-30: This sentence seems to repeat the one in Line 23-26. Please rewritten Line 22-30 to make the meaning clear and avoid overlap.

We highly appreciate these detailed comments which have helped us improve the paper quality a lot. We have specified the time period of the trend, and rewritten the Lines 22-30 as suggested, which are in Lines 14-30: "**During the observation period from 2001 to 2020, the annual aerosol optical depth (AOD) at most sites showed increasing trends (0.002-0.029 yr^{-1}) except for that at three sites of Canberra, Jabiru, and Lake Argyle, which showed decreasing trends (-0.005 - -0.002 yr^{-1}). In contrast, the annual Ångström exponent (AE) showed decreasing tendencies at most sites (-0.045 - -0.005 yr^{-1}). The results showed strong seasonal variations in AOD with high values in the austral spring and summer and relatively low values in the austral fall and winter, and weak seasonal variations in AE with the highest mean values in the austral spring at most sites. Monthly averaged AOD increases from August to December or next January, and decreases during the March-July. Spatially, the MODIS AOD showed obvious spatial heterogeneity with high values appeared over the Australian tropical savanna regions, Lake Eyre Basin, and southeastern regions of Australia, while low values appeared over the arid regions in western Australia. The MERRA-2 showed that carbonaceous over northern Australia, dust over central Australia, sulfate over densely populated northwestern and southeastern Australia, and sea salt over Australian coastal regions are the major types of atmospheric aerosols. The nine ground-based AERONET sites over Australia showed that the mixed type of aerosols (biomass burning and dust) are dominant in all seasons. Moreover, the CALIPSO showed that polluted dust is the dominant aerosol type detected at heights 0.5 - 5 km over Australian continent during all seasons. The results suggested that Australian aerosol has similar source characteristics due to the regional transport over Australia, especially for biomass burning and dust aerosols. However, the dust-prone characteristic of aerosol is more prominent over the central Australia, while the biomass burning-prone characteristic of aerosol is more prominent in northern Australia.**".

2. Section 2.2.1: Line 104-106: Does this affect the trend analysis? You may want to do a sensitivity analysis using level 1.5 data for all years and compare with the present results.

This is a good question. Following the suggestion, we performed a sensitivity analysis between AERONET Daily L1.5 and L2 AOD products, and found that this dataset issue does not affect the trend analysis. As shown in Figure R1, the AOD trends derived from the AERONET Daily L1.5 product are very similar to the trends estimated from combined of Daily L1.5 (2001-2018) and L2.0 (2019-2020) AERONET products (Figure 3 in the manuscript). We have added this information into the manuscript in Line 106-107: "**A sensitivity analysis showed that the use of level 1.5 data**

should not affect the trend analysis since they show the similar trends for period 2001-2018 when both datasets are available.”

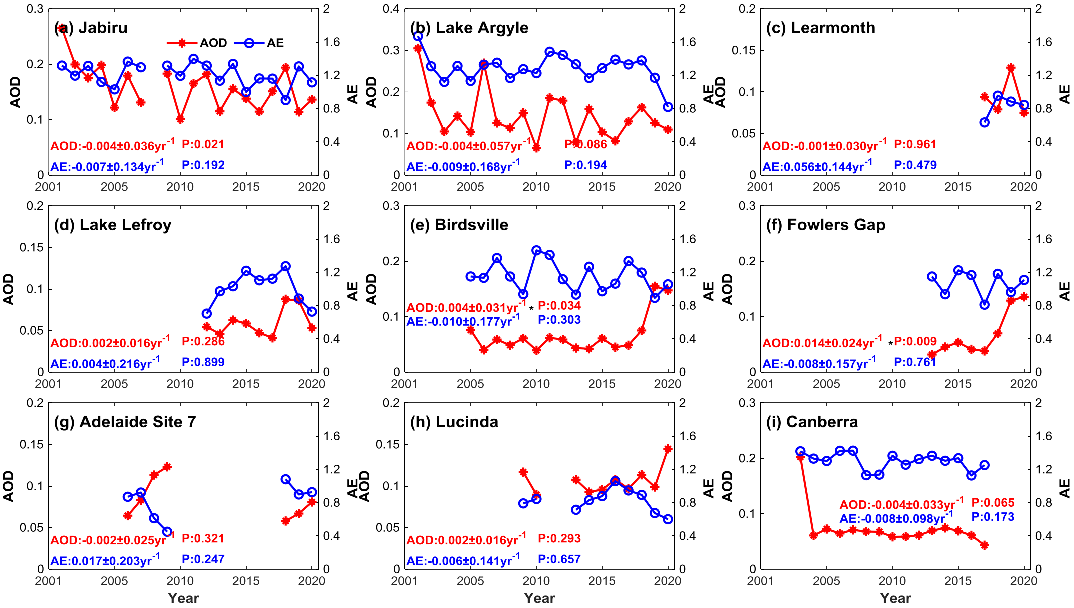

Figure R1. Temporal variations of annual mean AOD, AE (Level 1.5) at nine AERONET sites in Australia. Note: “*” means passing the confidence testing at α=0.05.

3. Section 2.3: Line 146-148: Clarify what data sources you used to determine the aerosol types.

We have clarified the data sources (i.e., AERONET) which are used to determine the aerosol types by using the two methods from Kaskaoutis et al. (2007) and Giles et al. (2012), which are in Lines 158-160: “**Two methods from Kaskaoutis et al. (2007) and Giles et al. (2012) are adopted to distinguish aerosol types by using the AOD at 500 nm, AE at 440-870 nm, and SSA at 440 nm which are derived from AERONET, as illustrated in Fig. 2.**”

4. Section 3.1.1: Whenever you describe a trend, please specify what time period the trend is for. A trend without a time period is difficult to interpret. Please also describe whether the trends are statistically significant or not. Also, please try to explain why AODs present increasing or decreasing trends at the nine sites. Line 166-167: It is not appropriate to use “trend” for only two years.

We highly appreciate these suggestions. The time period and significance of trend are now added in the revised manuscript. Moreover, we have tried our best to find and then describe the reasons for the increasing/decreasing trend at the nine sites. We have also removed the sentence regarding the trend for that two years since it is not appropriate. The corresponding changes are in Lines 171-181: “**It showed an increasing tendency in the annual mean AOD at most sites in Australia during the observation period from 2001 to 2020 except for the sites of Canberra, Jabiru, and Lake Argyle, at which the annual mean AOD showed a decreasing trend of -0.004±0.033 yr^{-1}, -0.002±0.035 yr^{-1}, -0.004±0.057 yr^{-1}, respectively. There were no statistically significant trends at most sites during the observation period from 2001 to 2020 except for the sites of Birdsville and Fowlers Gap. The general AOD decrease at sites over northern Australia and the general increase at sites over central Australia are consistent with the findings obtained by Mehta et al. (2016) using MODIS and Multi-angle Imaging SpectroRadiometer (MISR) AOD dataset during the period 2001–2014 and Mitchell et al. (2010) using site observations over the decade 1997–2007. The decreasing annual trends over northern Australia could be associated with the decreases in BC and OC AODs (Yoon et al.,2016).**

However, increasing AOD trends over central Australia could be mainly attributed to the increase of dust activities (Mitchell et al.,2010)." and in Lines 185-188: "**It was worth mentioning that significant increases of AOD are observed during the period 2019-2020 at most sites, such as Adelaide Site 7, Birdsville, Fowlers Gap, and Lucinda. The increases in AOD are related to the frequent fire activities in Australia from September 2019 to January 2020.**"

5. Section 3.1.2: Please explain why AODs at the nine sites are generally high in spring and summer and low in fall and winter, before explaining why some sites peak in spring and others peak in summer. Line 230: What "marine biogenic emissions" are these? Are they sea-spray aerosols? I doubt whether "biogenic" is an appropriate term here. Line 237: I suggest you avoid using "trend" here. This is confusing since you used "trend" represent interannual trend in the last section.

We appreciate all of these comments, which are very helpful to us. We have modified our descriptions accordingly based on these comments. In addition, we have removed "marine biogenic emissions" (Line 230) and "trend" (Line 237) since they are not appropriate as the reviewer questioned. The new descriptions are now in Lines 237-259 as follows: "**The main contributors to the high AODs in spring and summer were smoke emissions from biomass burning, dust storms, marine emissions from the sea spray produced in breaking waves, and the natural sulfate particles released from phytoplankton (Rotstayn et al., 2010). The occurrence frequency and intensity of dust storm activities and wildfires decreased in fall and winter, resulting in low AOD values in those two seasons over Australia. Furthermore, the highest seasonal average AOD values were observed in spring at Birdsville (0.09), Jabiru (0.22), Lake Argyle (0.23), and Lucinda (0.13), while they were observed in summer at the other five sites (0.07-0.11). The seasonal variations in AOD observed by AERONET and MODIS were in good agreement at nine sites apart from small differences in magnitude. Mitchell et al. (2013) also found that the AOD values at Wagga and Canberra peaked in summer, while AOD values peaked in spring at sites that are located in the arid zone. This is due to the increasingly forested and bushfire-prone characteristics at the more easterly sites (Mitchell et al., 2013). Moreover, less precipitation and higher wind speeds during spring were observed in northern Australia (north of 18°S), which may lead to the increase in AOD from biomass burning and long-range transport of marine emissions (Fig. 8). During summer, the decrease in AOD was significant over northwestern regions, consistent with the large increase in precipitation and decrease in wind speeds. However, the increase in AOD in eastern and southeastern regions during summer could be associated with the increase of biomass burning and sea salt aerosols that were transported from the Pacific Ocean.**

The seasonal variation in AE was different from that in AOD. There were no obvious seasonal variations in AE at the nine sites. The maximum seasonal mean AE values (0.92-1.43) were observed in spring at all sites except for Canberra, which was mostly related to the fine particles from biomass burning in spring. Similar results were reported by Mitchell et al. (2013) during the period 1998-2012 in northern Australia. Further, the seasonal mean AE values were greater than 1.0 over all seasons at Canberra, Jabiru, Lake Argyle, Fowlers Gap, and Birdsville, while the mean AE values were less than 1 over all seasons at the Adelaide Site 7 and Luncinda. In addition, at Learmonth and Lake Lefroy, high AE values (0.98-1.07) were observed in spring and fall, and low values (0.56-0.99) were observed in summer and winter."

6. Section 3.2.2: I think a lot of descriptions in the first paragraph are based on Figure 15, but

this figure is not cited until the next paragraph. Line 246: Is it really "intercontinental transport"? I think the sentences below do not actually talk about transport from other continents.

The reviewer proposed good questions. We agree with the reviewer and have changed our descriptions by expressing the corresponding figures for our descriptions for the whole section.

It is actually not "intercontinental transport" - we used wrong word since what we want to express is the "long-range transport". We have deleted the wrong description and modified our descriptions at Lines 386-389: "**Figure 15i and Figure S5 showed that air masses originated mainly from the Indian Ocean (>50%), crossing the regions that are affected by wildfires during the spring and summer season, and reaching the Canberra site, which indicated a possible transport of biomass burning and clean marine aerosols from forest regions and ocean, respectively.**".

7. Section 3.2.3: Please comment on whether the distribution of MERRA-2 based aerosol compositions are consistent with the AERONET based aerosol types.

We appreciate this suggestion. We add a description about this at Lines 391-393: "**We should note that the AERONET-based aerosol types show similar spatial distributions as follows by using the MERRA-2 based aerosol types, while they are slightly different in aerosol types classified.**"

8. Section 3.3: Whenever possible, please try to make connections between the CALIPSO based aerosol types and the AERONET based aerosol types used in previous sections. Second paragraph: please clarify that this paragraph describes the vertical profiles averaged across Austria, not one of the three regions defined in the last paragraph.

We appreciate these suggestions and made changes to Section 3.3 accordingly. First, the descriptions about the connections between the CALIPSO based aerosol types and the AERONET based aerosol types are added:

Lines 453-455: "**Further, the aerosol types classified by AERONET over Australia also indicated that the mixed type of aerosols (mostly biomass burning and dust) is the dominant type during all seasons. The result suggested the significant impacts of both biomass burning and desert emissions in Australia.**"

Lines 472-476: "**The results confirmed the existence of dust aerosols in northern Australia, which were mostly generated along with fires and transported from south inland deserts. The altitude with peak occurrence frequency (~5%) for elevated smoke was ~3 km throughout the year. Higher occurrence frequency of elevated smoke was observed at heights from 2 to 4 km in spring, consistent with the result of prevalence of biomass burning aerosol in spring at Lake Argyle, which is located in the biomass burning regime.**"

Second, we clarified that the average vertical profile values are for whole Australia (i.e., the blue shade area in Fig. 18) in Lines 448-449: "**Fig. 19 shows the averaged occurrence frequency profile of each aerosol type in each season from 15-year CALIPSO observations in whole Australia (blue shade region in Fig. 18).**".

9. Figure 15: Clarify that the aerosol types are derived from AERONET

We have revised the caption of Figure 15 (Figure 14 in revised manuscript) to Clarify that the aerosol types are derived from AERONET: "**Relative percentages of different aerosol components in each season at nine sites which are derived from AERONET during the observation period, including dust, biomass burning, mixed, urban/industrial, and uncertain aerosol types.**"