Reply to Anonymous Reviewer #3:

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. I have a couple of questions regarding the data. In Line 105, I understand that Level 1.5 data from AERONET has much large volume. But it also shows higher uncertainty I wonder if any quality control on this data is performed? Actually, keeping all quality screen criteria from AERONET Level 2 algorithm except the AOD threshold will keep most of the data.

The reviewer proposed a good question. Indeed, Level 1.5 data from AERONET has relatively large uncertainty. Thus, we used level 2.0 data covering most study period (i.e., 2001-2018). However, only five AERONET sites (i.e., Lake Lefory, Learmonth, Jabiru, Lake Argyle, Lucinda), which were located at Australian coastal region, have level 2.0 quality controlled and cloud screened data in 2019. Moreover, only one AERONET site has level 2.0 data in 2020. Hence, there is a lack of level 2.0 data for the central Australia. To obtain a long time series of ground-based observations and to analyze the spatial and temporal characteristics of aerosols over Australia in combination with remote sensing and reanalysis data, we used level 1.5 data for the period 2019-2020 along with the level 2.0 data for the period 2001-2008.

To make sure whether this combination will affect our trend analysis, 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.**".

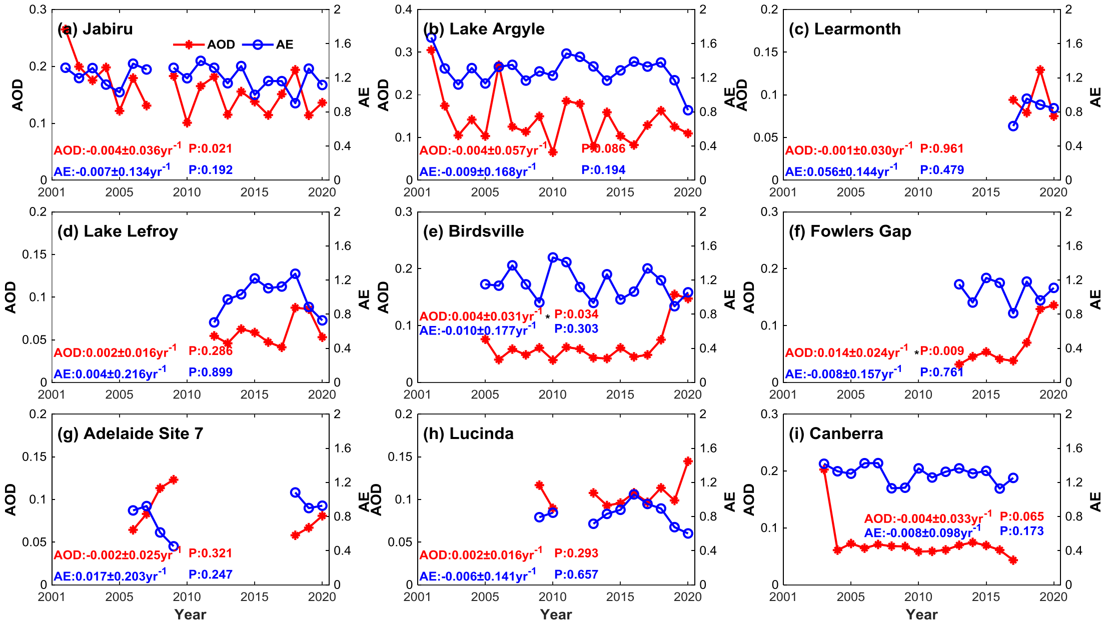

Figure R1. Temporal variations of annual mean AOD, AE (Level 1.5) at nine AERONET sites in Australia. Note: "*" means passing the confidence testing at α=0.05.

2. Section 2.2.2: is there a particular reason that MODIS Terra AOD is not used?

The reviewer proposed a good question. The reason that we only use one satellite AOD product is that they provide basically the same (very similar) long-term statistical characteristics of AOD (Sayer et al., 2015). Actually, Sayer et al. (2013) showed that AOD errors compared to AERONET appear to be slightly larger for Terra than those from MODIS Aqua by around 3% of the AOD. Considering that we are carrying statistical analysis instead of short-term event analysis, we did not use the MODIS Terra AOD. Of course, when studying short-term AOD characteristics or pollution events, adding MODIS Terra AOD would be much helpful.

Sayer, A. M., Hsu, N. C., Bettenhausen, C., and Jeong, M.-J.: Validation and uncertainty estimates for MODIS Collection 6 "Deep Blue" aerosol data, Journal of Geophysical Research: Atmospheres, 118, 7864-7872, https://doi.org/10.1002/jgrd.50600, 2013.

Sayer, A. M. , Hsu, N. C. , Bettenhausen, C. , Jeong, M. J. , & Meister, G. . (2015). Effect of modis terra radiometric calibration improvements on collection 6 deep blue aerosol products: validation and terra/aqua consistency. Journal of Geophysical Research: Atmospheres.

3. Section 2.2.3: the aerosol type analysis is primarily based on MERRA -2 data. I wonder if there is any validation of the MERRA 2 aerosol types, considering that there can be uncertainties in the model simulations?

This is a good question. It is really challenging for us to evaluate the aerosol types from MERRA-2 without sufficient information. However, considering the potential uncertainties in MERRA-2 data, we have also carried out the aerosol type analysis in Australia by using the combination of AERONET and CALIPSO data in section 3.3, which show very similar results in the spatial distributions of aerosol types and might imply the reliability of the aerosol types from MERRA-2 indirectly.

4. Section 3.1.1: the significance level of all trends should be provided here, especially that most trends are rather small.

We agree with the reviewer. We added significance level information into Figure 3 and revised the corresponding descriptions in Section 3.1.1 in the modified manuscript. More details please see the revised manuscript in Section 3.1.1.

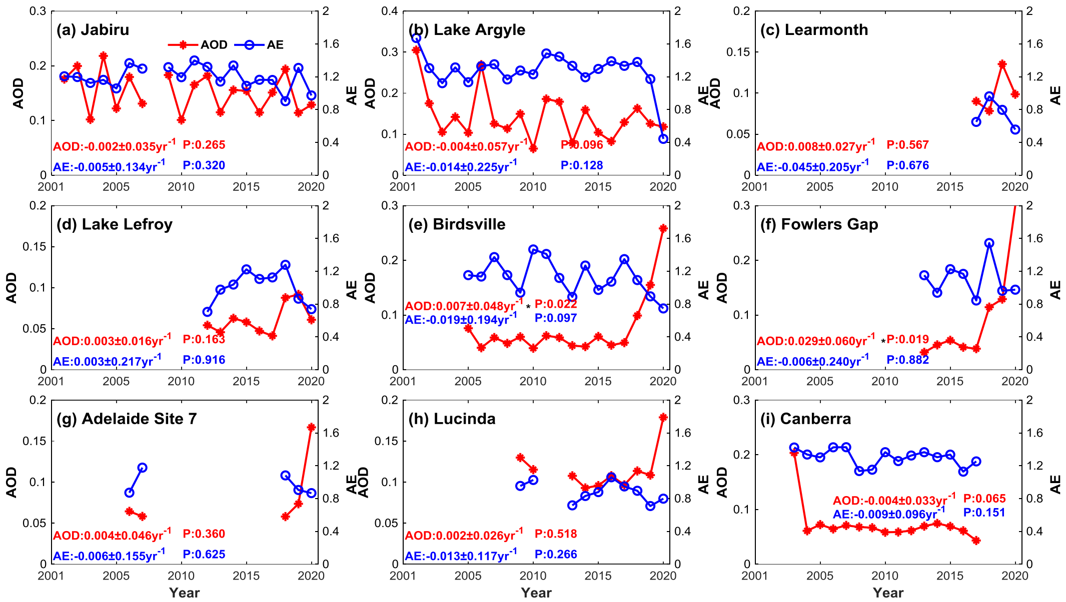

Figure 3. Temporal variations of annual mean AOD, AE at nine AERONET sites in Australia. Note: "*" means passing the confidence testing at α=0.05. "P" means P value.

5. 3.2.2: I have two questions here. First, I wonder how the back trajectories are clustered? Which method is used? Is it subjective or objective? Second, I think the aerosol source analysis needs to be combined with aerosol type analysis, i.e., what are the potential sources of each aerosol type at each site? For this purpose, I suggest the authors separate the trajectory analysis by aerosol type or by season, according to the results of Figure 15.

We appreciate these questions. In this study, we first used the Python + HYSPLIT to generate trajectories during the study period (i.e.,2005-2020). Then we employed the TrajStat module from Meteoinfo version2.4.1 to cluster the back trajectories (http://meteothink.org/docs/trajstat/cluster_cal.html). There are two clustering options with Euclidean distance or angle distance. In this study, we used the Euclidean distance method for cluster Calculation. Moreover, the Total spatial variation (TSV) was calculated to determine the class number of back trajectories. Thus, the cluster method is objective. The TSV percent change vs cluster numbers figure can indicate the suitable cluster number before dramatic increasing of TSV percent. More detail information please see the introduction of the TrajStat module (http://meteothink.org/docs/trajstat/cluster_cal.html). We have briefly indicated the cluster method in our method part in Section 2.3 at Lines 166-167: "Note that we have employed the TrajStat module from Meteoinfo version2.4.1 to cluster the back trajectories by using the Euclidean distance method (http://meteothink.org/docs/trajstat/index.html)."

Second, we have separated the trajectories by season based on your suggestion, with the Figures shown in the supplementary (Figs. S2-S5). Following your suggestion, we have added the relevant conclusions in our revised manuscript at several parts. For example,

Line 344-348: "**Furthermore, the seasonal trajectories evidenced the dust aerosols transport from the southeastern deserts (Fig. S2). The results were similar to the findings of McGowan and Clark (2008), who also demonstrated that dust transport from Lake Eyre could travel through the northwest dust transport pathway to affect the northern Australia, Indonesia and the southern Philippines by using HYSPLIT model during 1980-2000.**".

Line 369-374: “**Seasonal trajectories showed that 50% of the airflow at Birdsville was from the eastern and southeastern Australia during four seasons, which evidenced the biomass burning aerosols transport, especially during spring and summer (Fig. S4). This was supported by the findings of Qin et al. (2009), who demonstrated that who demonstrated that smoke generated from fires in Canberra could be transported over 1500 km across New South Wales to the central Australia.**”.

Line 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.**”.

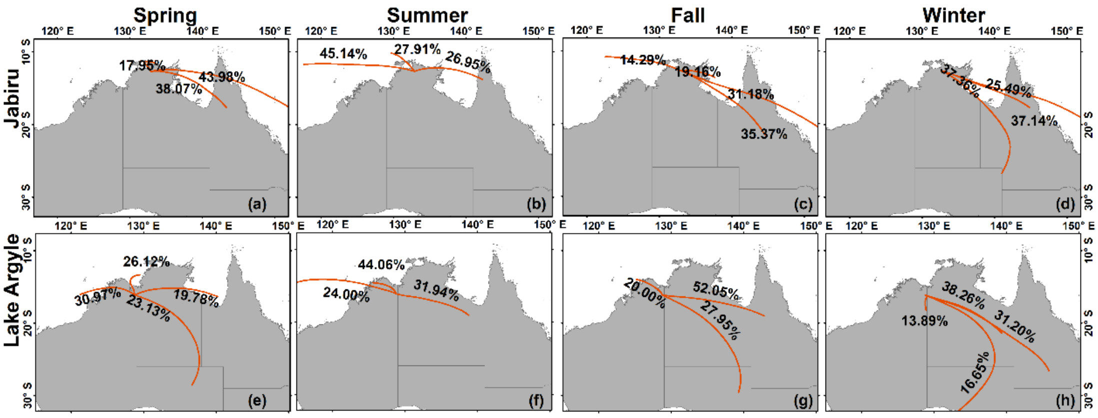

Figure S2. Cluster analysis of simulated back trajectories from HYSPLIT during the period January 2005-May 2020 for air masses ending at Jabiru and Lake Argyle at 500 m above ground level in Australia.

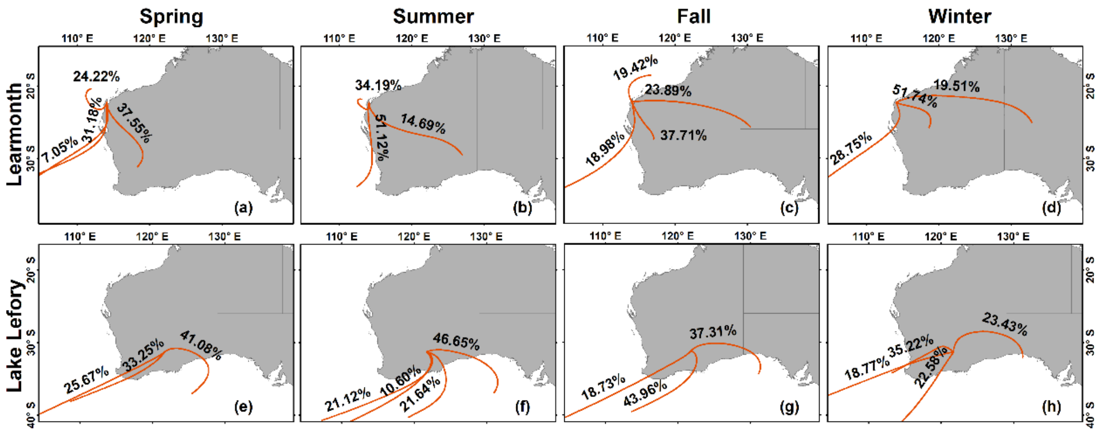

Figure S3. Cluster analysis of simulated back trajectories from HYSPLIT during the period January 2005-May 2020 for air masses ending at Learmonth and Lake Lefory at 500 m above ground level in Australia.

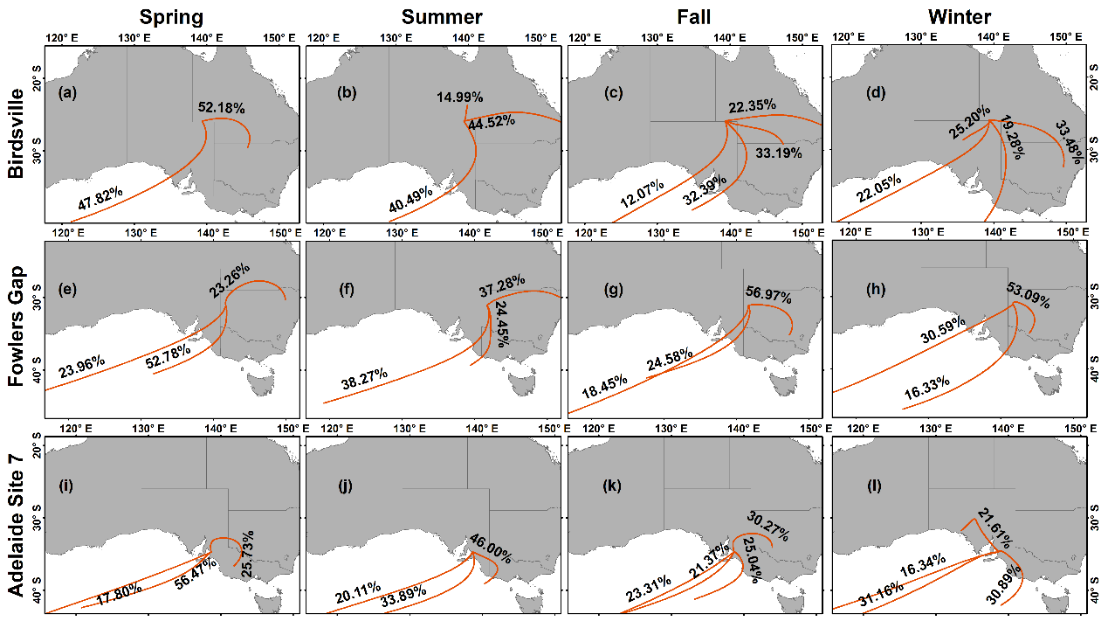

Figure S4. Cluster analysis of simulated back trajectories from HYSPLIT during the period January 2005-May 2020 for air masses ending at Birdsville, Fowlers Gap, and Adelaide Site 7 at 500 m above ground level in Australia.

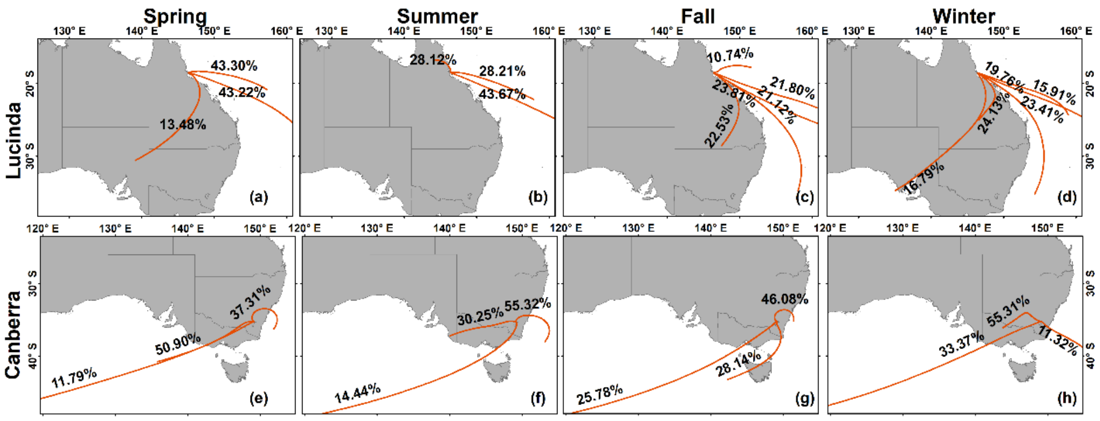

Figure S5. Cluster analysis of simulated back trajectories from HYSPLIT during the period January 2005-May 2020 for air masses ending at Lucinda and Canberra at 500 m above ground level in Australia.

6. I am curious about how aerosol properties change during the extremely intense wildfire in late 2019/early 2020. It seems that AOD has greatly increased over Victoria and Australian Capital Territory. Did the authors see other changes in aerosol properties, e.g., AE, absorption, aerosol type, etc? Btw, the location of Australian Capital Territory is not marked on Figure 1.

The reviewer proposed a very good point. Yes, we did see the changes in aerosol properties during this extreme intense wildfire event. The extremely intense wildfire during the 2019/2020 fire season had significant impact on aerosol properties, such as the extreme increase in AOD for most southeastern Australia, the dominance of fine particle aerosols, and the significant increase in carbonaceous and dust aerosols in southeastern and central Australia, respectively. We are making a comprehensive special investigation about the optical and physical properties of aerosols during the Australia wildfires in 2019/2020 following this study, and would be more than happy to provide you those results in future.

In Figure 1, we now add the label of Australian Capital Territory.

7. My final comment is that there is lack of a comparison with previous studies. What are the major new findings of the current study as compared with previous studies on aerosol properties in Australia? Is the analysis of aerosol type in this study supported by previous in-situ measurements? Some discussion should be added.

We appreciate this valuable comment. In most of previous studies, fire season has often been the focus study period due to the significant impacts of biomass burning aerosol. Meanwhile, most of previous studies have focused on aerosol properties at a specific site/region or short-term variations of aerosols due to the difficulty of obtaining ground-based aerosol data. This study analyzed the long-term spatial and temporal properties of aerosols over Australia from ground-based measurements (i.e. AERONET), remote sensing observations (MODIS, CALIPSO), and reanalysis data. The major new findings are about the long-term trends of AOD and AE, the spatio-temporal variations of AOD, and the vertical distributions of aerosol optical properties.

Following this suggestion, we have added the discussions by comparing with previous findings in our revised manuscript at several parts. For example,

Line 342-343: "**Karlson et al. (2014) analyzed the element and particle size of dust deposited in northwestern Australia from 2008 to 2009 and found that samples of Halls Creek (located in the southwest of lake argyle) were derived from the central Australia (e.g., the Lake Eyre Basin).**".

Line 371-373：“**This was supported by the findings of Qin et al. (2009), who demonstrated that smoke generated from fires in Canberra could be transported over 1500 km across New South Wales to the central Australia.**”

Line 382-384: "**Many previous studies also showed that urban/industrial aerosols and biomass burning aerosols were the main components of aerosols at Canberra (Qin et al., 2009; Mitchell et al., 2006; Provençal et al., 2017). Moreover, clean marine and dust aerosols were abundant during fall and winter, which was associated with its location in coastal areas and southeast dust transport corridors (McGowan and Clark., 2008).**".