## **Reply to anonymous referee #1:**

 We appreciate the reviewer for his/her thoughtful comments and suggestions, which are very helpful in improving our manuscript. We have carefully considered all the comments and revised the manuscript and the supplementary accordingly. Below is a point-by-point response to these comments.

- **Minor comments:**
- 1. General question about the stations selected in this study: I'm curious to know why polar stations are not included in the analysis, despite some of them, such as Opal or Andenes, having long-term observations. It is also evident from Figure 1 that 10 there is a clear bias towards Europe and the United States.

 The stations in this study is selected based on the method described in Sect. 2.1, which requires stations to have at least 8 monthly measurements for each year for temporal representativeness. Polar stations often have no monthly measurements in winter due to inadequate sunlight and thus fail to meet the above condition. Fig. R1 shows the monthly median AOD for two polar sites, OPAL and Andenes. The two sites have no data during November-February, and the time series for OPAL is much discontinuous. Considering that the measurements at polar stations concentrate in fixed seasons (summer), we revised the standard for polar stations in the MS (lines 95-97):

 *"Considering polar stations often have no monthly measurements in winter, the least number of monthly medians for each year are reduced to 4 for stations at latitudes above 65 degrees."*

- Seven sites (Andenes, Barrow, Hornsund, Kangerlussuaq, PEARL, Resolute\_Bay, and Thule) located in the Arctic was selected in this study, and all of them exhibit negative AOD trends, suggesting decreased AOD in the Arctic.
- Indeed, the selected stations are biased towards North America and Europe. 61 and 48 stations selected in this work are located in Europe and North America respectively. This is due to the higher density and better data maintenance of the stations in Europe.



**Figure R1: Time series of 440 nm AOD at (a) OPAL and (b) Andenes.** 

 2. Figure 2 and the stations selected for discussion: The rationale for selecting the stations displayed in this figure is not clear.

 Fig. 2 is only a reference for reader to know the location of stations mentioned in the MS, which has been mentioned in the MS in line 105:

*"Locations of stations mentioned in the manuscript are presented in Fig. 2."*

 These stations are mentioned when analyzing particular cases (i.e., Birdsville), or showing time series as representative stations at specific regions. For the latter, the stations are selected according to their spatial representativeness, length of records, and significance

and magnitude of the trends for some parameters.

 Locations of other stations could be found in the supplementary, which has been mentioned in the MS in line 103:

*"Locations, trends and time series for all the stations could be found in the supplementary."*

 3. General comment about general-global trends results: Given that this paper aims to study general trends on a global scale, I wonder if it would be more appropriate to quantify the results in terms of regions. Currently, the quantification of the observed trends is done only in terms of the different stations defined (in a way that is not clear to me) in Figure 2.

 Thanks for the suggestion. We did attempt to calculate regional trends. However, considering the lifetime and spatial heterogeneity of aerosols, the ground-based stations have limits in spatial coverage and representativeness, and for some regions, the numbers of stations are too few to represent the entire region. Moreover, direction of regional trends could be summarized qualitatively if the trends are coherent for stations in the region, but it is difficult to quantify the magnitude and significance of the trends for a region, as trends of some stations are not significant or even opposite to those of most stations in the region. Therefore, we mainly summarized the magnitude and significance of the trends for the majority of the stations in a specific region.

 As mentioned in Minor Comment #2, Fig. 2 is only a reference for the readers to know the location of stations mentioned in the MS. Only representative stations are marked. When there are very limited stations located in the region, we discuss the station-based trends. When there are many stations with coherent trends, we discuss the trend by region. We have also added a table summarizing the trends and locations of all stations in the supplementary, and mentioned it in the MS in line 103:

*"Locations, trends and time series for all the stations could be found in the supplementary."*

 4. A more general question: Do the authors have any ideas about the general lack of statistical significance of the results found over the African continent? While AOD and AE trends are significant over the Arabian Peninsula, suggesting a possible increase in dust activity in this region, there is no statistical significance over Africa. Some recent studies show declining DOD trends across the Sahara and the Eastern Mediterranean. Do the authors have any insights on this?

 Aerosols in West Africa are primarily composed of dust, which has strong natural variability, making it difficult to obtain a significant trend. Trends of dust loading in Sahara is still uncertain. Shao et al. (2013) reported decreased dust activities in Sahara, whereas Merdji et al. (2023) reported increased dust loading. Trends in the two studies are generally weak and not that significant. In our work, we also found the trends in aerosol parameters generally insignificant or spatially incoherent, as can be seen in the time series of stations, Banizoumbou and IER\_Cinzana. AOD and AE both exhibit substantial variability, ranging from 0.2 to 1.0, and the trends are weak and insignificant.



 **Figure R2: Left: Time series of 440 nm AOD. Right: Time series of 440-870 nm AE. (a, b) Banizoumbou, (c, d) IER\_Cinzana.**

- **Technical comments:**
- 1. Figure 1: Why is panel (a) labeled as "Solar" Level 2.0? I recommend using the 82 terminology "Direct Sun," consistent with AERONET products.
- Thanks for the suggestion. We have revised the title of Fig. 1(a) in the MS.
- 84 2. Line 96: I don't understand the relevance of mentioning the "unique data logging" system used in Australia. Was there a problem with the acquisition time of the photometer?

 We are sorry for the confusion. We found a jump in AOD (more than a doubling of AOD) at Birdsville in 2019 and 2020, which coincides with the timing of the update of the

 algorithm. This jump could also be found in Yang et al. (2021). This is likely due to a data filtering artifact of the QA of the algorithm of Giles et al. (2019) that eliminated only the low AOD days (personal communication, T. Eck). This particular issue involves the way data are uniquely time stamped in Australia and does not occur at sites in the rest of the network.

 3. Line 97: In line with the previous comment, the authors mention an unnatural increase in AOD in Birdsville. Are the authors referring to a diurnal cycle or to the Kciclo, as explained by Cachorro et al. (2009) and subsequent papers?

 The jump of AOD at Birdsville could be observed on monthly and annual time series. According to Cachorro et al. (2008), the difference caused by KCICLO seems to be largely reduced when analyzing monthly and annual averaged data. We tend to believe that this discontinuity was caused by the algorithm upgrade. When upgrading the algorithm in the future to V4 of the AERONET database, this problem might be solved (personal communication, T. Eck).

103 4. Line 120: Sea salt is not included in the aerosol typing, even though it is one of the most abundant aerosol species in Earth's atmosphere, and its hygroscopicity is an important parameter for quantifying its interaction with solar radiation.

 We are sorry for the confusion. We also think that sea salt has important climate effect. In this study, sea salt is only excluded in aerosol type analysis (Sect. 3.3), because this type accounts for only 2.5% percent of total records which is too small to calculate trends, and is mainly detected at oceanic stations with low AOD levels and thus high uncertainties. When analysing AOD, AE, AAOD, and SSA, sea salt records are not excluded. We have revised the description in lines 170-174 for clarity:

<sup>112</sup> *"It should be noted that sea salt aerosols typically having FMF*<sub>550</sub> *below 0.4 and SSA*<sub>440</sub> *around 0.98 (included in the "Uncertain" type in Table 1) are not considered in the analysis of aerosol type trends (Sect. 3.3), because most AERONET stations are located over land where sea salt is not the predominant type, and sea salt aerosols only account for a negligible proportion (about 2.5% for "Uncertain" type)."*

 5. Figure 3: I find this figure (and the following figures that use the same criterion) difficult to understand due to the exclusive use of dots. I suggest that the authors improve the figure by using different symbols to indicate varying levels of 120 statistical significance.

 Thanks for the suggestion. We updated the maps in the MS with different symbols to indicate differnet levels of statistical significance. Specifically, we use dots to indicate trends at 90% significance, and use triangle to represent trends below 90% significance level.

125 6. Figure 4: Are the stations used in this figure selected for a specific reason? Are they chosen based on their geographical location, or do they represent significant trends? Additionally, why does Figure 4c contain two different stations in the same panel? It is difficult to distinguish between the two lines. Another suggestion is to include 129 the country name in each subfigure label to help focus the reader's attention on the specific region discussed in the text. This suggestion could also be applied to other 131 similar figures.

Thanks for the suggestion. We have added names of the regions in each subfigure.

As mentioned in Minor Comment #2, the stations in Fig. 5 (and following similar figures)

 are mainly selected according to the spatial representativeness of stations, length of records, and significance and magnitude of the trends.

 The two stations, Beijing and XiangHe, are combined for better comparation, as explained in the MS in lines 200-202:

 *"A comparation between AOD*<sup>440</sup> *time series of XiangHe and Beijing (Fig. 5c), two stations located very close to each other in East China, would further reveal that the substantial reduction of AOD*<sup>440</sup> *mainly occurred in the later years."*

 7. Line 135: The authors discuss the different rates of AOD reduction found in Western Europe compared to the values reported by Li et al. (2014). It would be very helpful if they could include the specific numbers found in that paper and also reference Figures 4h and 4g.

 Thanks for the suggestion. The AOD reduction rates reported by Li et al. (2014) in Western Europe were -0.1 per decade, while those in this work are generally -0.05 per decade. We have added these comparations in the MS in lines 185-186:

 *"The rates of AOD*<sup>440</sup> *reduction in western Europe (about -0.05 per decade) are not as substantial as those reported in Li et al. (2014), which was -0.1 per decade, suggesting a decelerated aerosol reduction rate in Europe in recent years."*

- 8. Lines 141-144: The authors state that, according to previous studies, a substantial reduction in AOD has occurred in the last decade. However, looking at Figure 4a, 153 for instance, I see a reduction in AOD over the entire period, starting from 2002.
- Did the authors analyse the presence of any breakpoints in these datasets?

 We are sorry for the confusion. We have updated the result, and records before 2009 at Chen-Kung\_Univ are filtered. In fact, at most stations over East Asia, the AOD first increased or remained stable, and then decreased. The AOD reduction over these stations manly occurred after 2008 (i.e., Osaka, Beijing, and XiangHe). We have also revised the description in lines 194-198:

*"However, the trend of AOD*<sup>440</sup> *in East Asia is not coherent throughout the period of 2000-*

*2022. According to the AOD*<sup>440</sup> *time series (Fig. 5a-c), AOD*<sup>440</sup> *increased in the early* 

*2000s, and decreased rapidly in the later years since around 2008, consistent with other* 

*regional aerosol trend studies (Eom et al., 2022; Gupta et al., 2022; Li, 2020; Lyapustin* 

*et al., 2011; Meij et al., 2012; Ramachandran et al., 2020; Ramachandran & Rupakheti,* 

*2022; Yoon et al., 2012)."*

 9. Lines 150 and 162: The authors mention results for "several oceanic island stations" in these two lines, while they also state that sea salt aerosols, the dominant species 168 at these sites, are not included in the analysis. Do they expect a bias in these sites because of this omission?

We are sorry for the confusion. The sea salt aerosols are only excluded in aerosol type

analysis in Sect. 3.3, which have been explained in the response to Minor Comment #4.

All of the AOD, AE, AAOD and SSA trend analyses in the MS include oceanic sites.

 As sea salt is the dominant aerosol type at oceanic sites, the positive AOD trends for these stations could be mainly attributed to increases of sea salt aerosols. We have also added the description about increased sea salt at these oceanic sites in the MS in lines 218-220:

 *"In addition to Nauru which exhibits significant positive AOD*<sup>440</sup> *trend, some other oceanic stations worldwide also exhibit positive AOD*<sup>440</sup> *trends, suggesting a widespread* 

*increase in oceanic aerosols, primarily sea salts. This result is consistent with Hsu et al.* 

*(2012) who also reported an increase in oceanic AOD."*

180 10. Line 155: Is the AOD trend 0.066 per decade according to Figure 4e?

Thanks for pointing this out. We are referring to the trend in Fig. 5e here, which should be

 0.062 instead of 0.066. We forgot to update the value in the previous MS. We have updated Fig. 5e and revised the trend value to 0.062.

184 11. Line 158: Is the AOD trend 0.166 per decade according to Figure 4f?

 We are sorry that this is the same issue as that in the last comment. We have updated Fig. 186 5f and revised the trend value to 0.167.

- 187 12. Line 161: In line 96, the authors attribute the problems in Birdsville to the logging system, but now they attribute it to a data screening anomaly. I don't understand either of these terms. I suspect there is a calibration problem (diurnal cycle or 190 Kciclo); can the authors confirm?
- 191 We are sorry for the confusion. As detailed in Technical Comment  $#2$ , the artifact of the QA of the algorithm eliminated the low AOD records, thus likely led to a jump in AOD.
- 193 13. Line 194: The discussion introduced here about significant positive trends in some places in Asia is interesting. Why not include one of these stations in Figure 6?

195 Thanks for the suggestion. In fact, Chen-Kung Univ (Fig. 6a in the previous MS draft) is one of these stations in the Taiwan Island, which exhibit significant positive AE trend in the previous MS draft. However, in the updated result, the AE trend over most of these Asia stations are not significant or coherent, therefore we revised the analysis in the MS in lines 235-238:

 *"East Asia exhibits no significant AE*440\_<sup>870</sup> *trends, indicating weak changes in the ratio of fine-mode and coarse-mode aerosols. Therefore, the great decrease of aersol loading in East Asia revealed in Fig. 4 might be related to similar reductions in both anthropogenic fine-mode aerosols and coarse-mode dust in these areas."*

- 14. Section 3.2: The two paragraphs starting at lines 241 and 256 are meant to provide 205 the results related to AAOD and SSA, respectively. However, these two variables are mixed throughout both paragraphs, making it difficult for the reader to follow 207 the discussion. I wonder if the authors could present these two pieces of information in a clearer manner.
- Thanks for the suggestion. We have revised the two paragraphs to separately discuss the 210 two parameters.

## 211 15. Line 271: The authors mention a positive SSA trend in Solar Village. However, in Figure 12d, there is a negative SSA trend of -0.034 per decade. Can the authors clarify this discrepancy?

- We are sorry for the confusion. The SSA trend in Solar Village is negative. We have revised the MS in lines 297-298:
- *"Negative SSA*<sup>440</sup> *trend for Solar\_Village (Fig. 13b) in the Arabian Peninsula is attributed to increases in absorbing dust aerosols."*
- 218 16. Section 3.3: I recommend using italics or quotation marks when referring to the different types of aerosols, such as "Mixture," "Dust," or "Non-absorbing," for example. I also suggest including the abbreviations SA, MA, and HA in the figure captions or somewhere in the text, since they were introduced in Table 1 (page 6).
- Thanks for the suggestion. We have used quotation marks to refer to aerosol types in the MS.
- 224 17. Section 4: This section is quite long and difficult to read. Rather than focusing on highlighting the most relevant results of this study, it seems to center on the differences observed with the paper published by Li et al. (2014). I recommend

 summarizing and streamlining this section to emphasize the important findings of 228 the authors.

 Thanks for the suggestion. We have revised Sect. 4 into a more concise expression. In particular, we shortened the comparison with Li et al. (2014) and added more recent references.

## **References**

- Cachorro, V. E., Toledano, C., Sorribas, M., Berjón, A., Frutos, A. M. de, & Laulainen, N.
- (2008). An "in situ" calibration‐correction procedure (KCICLO) based on AOD diurnal
- cycle: Comparative results between AERONET and reprocessed (KCICLO method) AOD‐
- alpha data series at el arenosillo, spain. *Journal of Geophysical Research: Atmospheres*,
- *113*(D2).<https://doi.org/10.1029/2007jd009001>
- Eom, S., Kim, J., Lee, S., Holben, B. N., Eck, T. F., Park, S.-B., & Park, S. S. (2022).
- Long-term variation of aerosol optical properties associated with aerosol types over east
- asia using AERONET and satellite (VIIRS, OMI) data (20122019). *Atmospheric Research*,

*280*, 106457.<https://doi.org/10.1016/j.atmosres.2022.106457>

- Gupta, G., Venkat Ratnam, M., Madhavan, B. L., & Narayanamurthy, C. S. (2022). Long- term trends in aerosol optical depth obtained across the globe using multi-satellite measurements. *Atmospheric Environment*, *273*, 118953. <https://doi.org/10.1016/j.atmosenv.2022.118953>
- Hsu, N. C., Gautam, R., Sayer, A. M., Bettenhausen, C., Li, C., Jeong, M. J., et al. (2012).
- Global and regional trends of aerosol optical depth over land and ocean using SeaWiFS
- measurements from 1997 to 2010. *Atmospheric Chemistry and Physics*, *12*(17), 8037–8053.
- <https://doi.org/10.5194/acp-12-8037-2012>
- Li, J. (2020). Pollution trends in china from 2000 to 2017: A multi-sensor view from space.
- *Remote Sensing*, *12*(2), 208.<https://doi.org/10.3390/rs12020208>
- Li, J., Carlson, B. E., Dubovik, O., & Lacis, A. A. (2014). Recent trends in aerosol optical
- properties derived from AERONET measurements. *Atmospheric Chemistry and Physics*,
- *14*(22), 12271–12289.<https://doi.org/10.5194/acp-14-12271-2014>
- Lyapustin, A., Smirnov, A., Holben, B., Chin, M., Streets, D. G., Lu, Z., et al. (2011).
- Reduction of aerosol absorption in beijing since 2007 from MODIS and AERONET.
- *Geophysical Research Letters*, *38*(10), L10803.<https://doi.org/10.1029/2011gl047306>
- Meij, A. de, Pozzer, A., & Lelieveld, J. (2012). Trend analysis in aerosol optical depths
- and pollutant emission estimates between 2000 and 2009. *Atmospheric Environment*, *51*,
- 75–85.<https://doi.org/10.1016/j.atmosenv.2012.01.059>
- Merdji, A. B., Lu, C., Xu, X., & Mhawish, A. (2023). Long-term three-dimensional
- distribution and transport of saharan dust: Observation from CALIPSO, MODIS, and reanalysis data. *Atmospheric Research*, *286*, 106658. <https://doi.org/10.1016/j.atmosres.2023.106658>
- Ramachandran, S., & Rupakheti, M. (2022). Trends in physical, optical and chemical
- columnar aerosol characteristics and radiative effects over south and east asia: Satellite and
- ground-based observations. *Gondwana Research*, *105*, 366–387. <https://doi.org/10.1016/j.gr.2021.09.016>
- Ramachandran, S., Rupakheti, M., & Lawrence, M. G. (2020). Aerosol-induced
- atmospheric heating rate decreases over south and east asia as a result of changing content
- and composition. *Scientific Reports*, *10*(1).<https://doi.org/10.1038/s41598-020-76936-z>
- Shao, Y., Klose, M., & Wyrwoll, K. (2013). Recent global dust trend and connections to climate forcing. *Journal of Geophysical Research: Atmospheres*, *118*(19). <https://doi.org/10.1002/jgrd.50836>
- Yang, X., Zhao, C., Yang, Y., & Fan, H. (2021). Long-term multi-source data analysis
- about the characteristics of aerosol optical properties and types over australia. *Atmospheric*
- *Chemistry and Physics*, *21*(5), 3803–3825.<https://doi.org/10.5194/acp-21-3803-2021>
- Yoon, J., Hoyningen-Huene, W. von, Kokhanovsky, A. A., Vountas, M., & Burrows, J. P.
- (2012). Trend analysis of aerosol optical thickness and ångström exponent derived from
- the global AERONET spectral observations. *Atmospheric Measurement Techniques*, *5*(6),
- 1271–1299.<https://doi.org/10.5194/amt-5-1271-2012>