## 1 **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.

- 6 **Minor comments:**
- 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
   there is a clear bias towards Europe and the United States.

11 The stations in this study is selected based on the method described in Sect. 2.1, which 12 requires stations to have at least 8 monthly measurements for each year for temporal 13 representativeness. Polar stations often have no monthly measurements in winter due to 14 inadequate sunlight and thus fail to meet the above condition. Fig. R1 shows the monthly 15 median AOD for two polar sites, OPAL and Andenes. The two sites have no data during 16 November-February, and the time series for OPAL is much discontinuous. Considering 17 that the measurements at polar stations concentrate in fixed seasons (summer), we revised 18 the standard for polar stations in the MS (lines 95-97):

19 "Considering polar stations often have no monthly measurements in winter, the least20 number of monthly medians for each year are reduced to 4 for stations at latitudes above

21 *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.



28

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

30 2. Figure 2 and the stations selected for discussion: The rationale for selecting the31 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:

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

35 These stations are mentioned when analyzing particular cases (i.e., Birdsville), or showing

36 time series as representative stations at specific regions. For the latter, the stations are

37 selected according to their spatial representativeness, length of records, and significance

38 and magnitude of the trends for some parameters.

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

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

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.

47 Thanks for the suggestion. We did attempt to calculate regional trends. However, 48 considering the lifetime and spatial heterogeneity of aerosols, the ground-based stations 49 have limits in spatial coverage and representativeness, and for some regions, the numbers 50 of stations are too few to represent the entire region. Moreover, direction of regional trends 51 could be summarized qualitatively if the trends are coherent for stations in the region, but 52 it is difficult to quantify the magnitude and significance of the trends for a region, as trends 53 of some stations are not significant or even opposite to those of most stations in the region. 54 Therefore, we mainly summarized the magnitude and significance of the trends for the 55 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:

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

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?

69 Aerosols in West Africa are primarily composed of dust, which has strong natural 70 variability, making it difficult to obtain a significant trend. Trends of dust loading in Sahara 71 is still uncertain. Shao et al. (2013) reported decreased dust activities in Sahara, whereas 72 Merdji et al. (2023) reported increased dust loading. Trends in the two studies are generally 73 weak and not that significant. In our work, we also found the trends in aerosol parameters 74 generally insignificant or spatially incoherent, as can be seen in the time series of stations, 75 Banizoumbou and IER Cinzana. AOD and AE both exhibit substantial variability, ranging 76 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.

- 80 **Technical comments:**
- Figure 1: Why is panel (a) labeled as "Solar" Level 2.0? I recommend using the
   terminology "Direct Sun," consistent with AERONET products.
- 83 Thanks for the suggestion. We have revised the title of Fig. 1(a) in the MS.
- 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?

97 The jump of AOD at Birdsville could be observed on monthly and annual time series. 98 According to Cachorro et al. (2008), the difference caused by KCICLO seems to be largely 99 reduced when analyzing monthly and annual averaged data. We tend to believe that this 100 discontinuity was caused by the algorithm upgrade. When upgrading the algorithm in the 101 future to V4 of the AERONET database, this problem might be solved (personal 102 communication, T. Eck).

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:

112 "It should be noted that sea salt aerosols typically having  $FMF_{550}$  below 0.4 and  $SSA_{440}$ 113 around 0.98 (included in the "Uncertain" type in Table 1) are not considered in the analysis 114 of aerosol type trends (Sect. 3.3), because most AERONET stations are located over land 115 where sea salt is not the predominant type, and sea salt aerosols only account for a 116 negligible proportion (about 2.5% for "Uncertain" type)." 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
statistical significance.

121 Thanks for the suggestion. We updated the maps in the MS with different symbols to 122 indicate differnet levels of statistical significance. Specifically, we use dots to indicate 123 trends at 90% significance, and use triangle to represent trends below 90% significance 124 level.

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
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
similar figures.

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

133 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.

136 The two stations, Beijing and XiangHe, are combined for better comparation, as explained137 in the MS in lines 200-202:

138 "A comparation between  $AOD_{440}$  time series of XiangHe and Beijing (Fig. 5c), two 139 stations located very close to each other in East China, would further reveal that the 140 substantial reduction of  $AOD_{440}$  mainly occurred in the later years."

141 7. Line 135: The authors discuss the different rates of AOD reduction found in
142 Western Europe compared to the values reported by Li et al. (2014). It would be
143 very helpful if they could include the specific numbers found in that paper and also
144 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:

148 "The rates of  $AOD_{440}$  reduction in western Europe (about -0.05 per decade) are not as 149 substantial as those reported in Li et al. (2014), which was -0.1 per decade, suggesting a 150 decelerated aerosol reduction rate in Europe in recent years."

- 151 8. Lines 141-144: The authors state that, according to previous studies, a substantial
  152 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.
- 154 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<sub>440</sub> in East Asia is not coherent throughout the period of 20002022. According to the AOD<sub>440</sub> time series (Fig. 5a-c), AOD<sub>440</sub> 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)."

166 9. Lines 150 and 162: The authors mention results for "several oceanic island stations"
167 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
169 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.

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

173 As sea salt is the dominant aerosol type at oceanic sites, the positive AOD trends for these 174 stations could be mainly attributed to increases of sea salt aerosols. We have also added 175 the description about increased sea salt at these oceanic sites in the MS in lines 218-220:

176 "In addition to Nauru which exhibits significant positive AOD<sub>440</sub> trend, some other oceanic stations worldwide also exhibit positive AOD<sub>440</sub> trends, suggesting a widespread 177 178

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

179 (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?

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

182 0.062 instead of 0.066. We forgot to update the value in the previous MS. We have updated 183 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?

185 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 188 system, but now they attribute it to a data screening anomaly. I don't understand 189 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&#3, the artifact of 192 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 194 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 196 one of these stations in the Taiwan Island, which exhibit significant positive AE trend in 197 the previous MS draft. However, in the updated result, the AE trend over most of these 198 Asia stations are not significant or coherent, therefore we revised the analysis in the MS in 199 lines 235-238:

200 "East Asia exhibits no significant  $AE_{440_{-870}}$  trends, indicating weak changes in the ratio 201 of fine-mode and coarse-mode aerosols. Therefore, the great decrease of aersol loading in 202 East Asia revealed in Fig. 4 might be related to similar reductions in both anthropogenic 203 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
  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
  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 thetwo parameters.

## 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 revisedthe MS in lines 297-298:
- 216 "Negative SSA<sub>440</sub> trend for Solar\_Village (Fig. 13b) in the Arabian Peninsula is attributed
  217 to increases in absorbing dust aerosols."
- 218 16. Section 3.3: I recommend using italics or quotation marks when referring to the
  219 different types of aerosols, such as "Mixture," "Dust," or "Non-absorbing," for
  220 example. I also suggest including the abbreviations SA, MA, and HA in the figure
  221 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 theMS.
- 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 ofthe 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.

## 232 **References**

- 233 Cachorro, V. E., Toledano, C., Sorribas, M., Berjón, A., Frutos, A. M. de, & Laulainen, N.
- 234 (2008). An "in situ" calibration-correction procedure (KCICLO) based on AOD diurnal
- 235 cycle: Comparative results between AERONET and reprocessed (KCICLO method) AOD-
- alpha data series at el arenosillo, spain. Journal of Geophysical Research: Atmospheres,
- 237 *113*(D2). https://doi.org/10.1029/2007jd009001
- 238 Eom, S., Kim, J., Lee, S., Holben, B. N., Eck, T. F., Park, S.-B., & Park, S. S. (2022).
- 239 Long-term variation of aerosol optical properties associated with aerosol types over east
- asia using AERONET and satellite (VIIRS, OMI) data (20122019). *Atmospheric Research*,
- 241 280, 106457. https://doi.org/10.1016/j.atmosres.2022.106457
- Gupta, G., Venkat Ratnam, M., Madhavan, B. L., & Narayanamurthy, C. S. (2022). Longterm 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).
- 247 Global and regional trends of aerosol optical depth over land and ocean using SeaWiFS
- 248 measurements from 1997 to 2010. *Atmospheric Chemistry and Physics*, 12(17), 8037–8053.
- 249 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.
- 251 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
- 253 properties derived from AERONET measurements. Atmospheric Chemistry and Physics,
- 254 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).
- 256 Reduction of aerosol absorption in beijing since 2007 from MODIS and AERONET.
- 257 Geophysical Research Letters, 38(10), L10803. https://doi.org/10.1029/2011gl047306
- 258 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,
- 260 75–85. https://doi.org/10.1016/j.atmosenv.2012.01.059
- 261 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
- 265 Ramachandran, S., & Rupakheti, M. (2022). Trends in physical, optical and chemical
- 266 columnar aerosol characteristics and radiative effects over south and east asia: Satellite and
- 267 ground-based observations. Gondwana Research, 105, 366–387.
  268 https://doi.org/10.1016/j.gr.2021.09.016
- 269 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
- 271 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
- 275 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*
- 277 Chemistry and Physics, 21(5), 3803–3825. https://doi.org/10.5194/acp-21-3803-2021

- 278 Yoon, J., Hoyningen-Huene, W. von, Kokhanovsky, A. A., Vountas, M., & Burrows, J. P.
- 279 (2012). Trend analysis of aerosol optical thickness and ångström exponent derived from
- 280 the global AERONET spectral observations. Atmospheric Measurement Techniques, 5(6),
- 281 1271–1299. https://doi.org/10.5194/amt-5-1271-2012