

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 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.

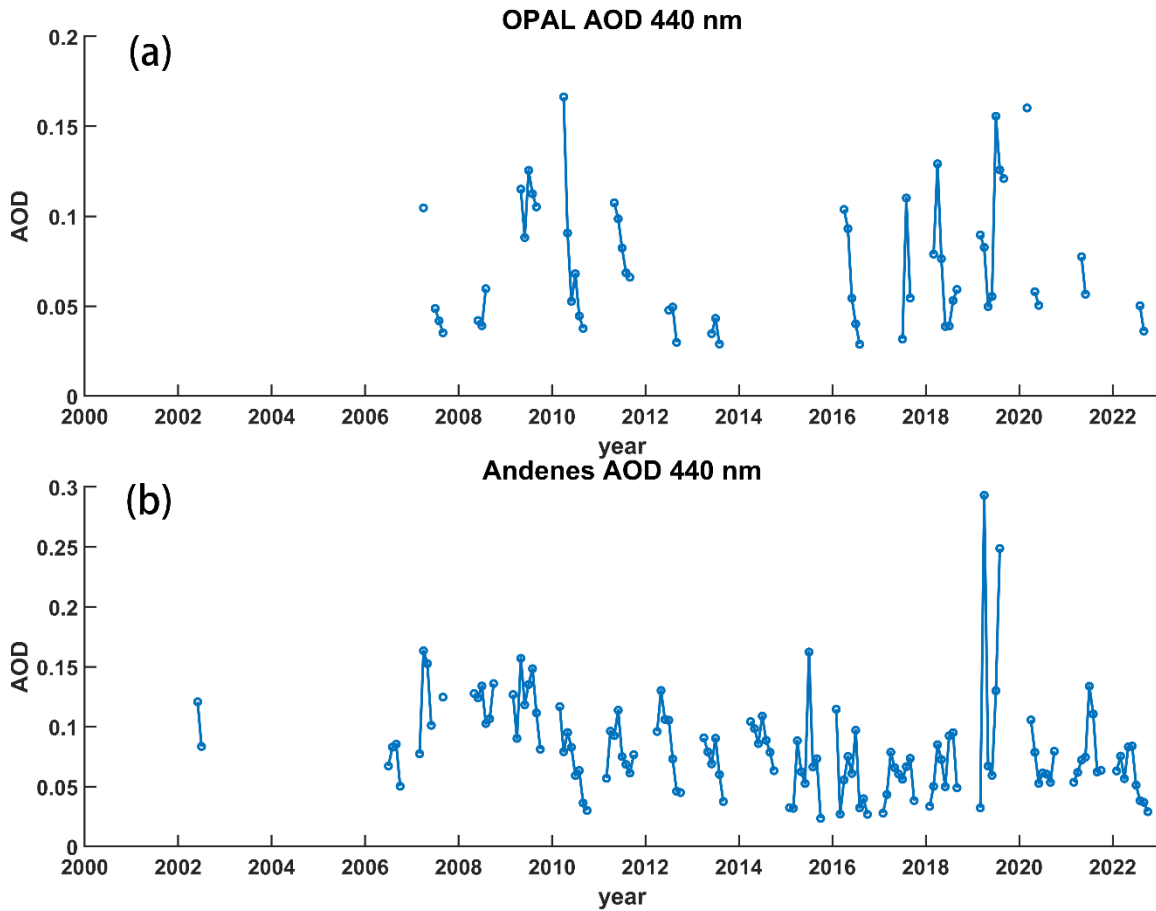


Figure R1: Time series of 440 nm AOD at (a) OPAL and (b) Andenes.

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:

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

- 42 3. General comment about general-global trends results: Given that this paper aims to
43 study general trends on a global scale, I wonder if it would be more appropriate to
44 quantify the results in terms of regions. Currently, the quantification of the observed
45 trends is done only in terms of the different stations defined (in a way that is not
46 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.

56 As mentioned in Minor Comment #2, Fig. 2 is only a reference for the readers to know the
57 location of stations mentioned in the MS. Only representative stations are marked. When
58 there are very limited stations located in the region, we discuss the station-based trends.
59 When there are many stations with coherent trends, we discuss the trend by region. We
60 have also added a table summarizing the trends and locations of all stations in the
61 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.”*

- 63 4. A more general question: Do the authors have any ideas about the general lack of
64 statistical significance of the results found over the African continent? While AOD
65 and AE trends are significant over the Arabian Peninsula, suggesting a possible
66 increase in dust activity in this region, there is no statistical significance over Africa.
67 Some recent studies show declining DOD trends across the Sahara and the Eastern
68 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.

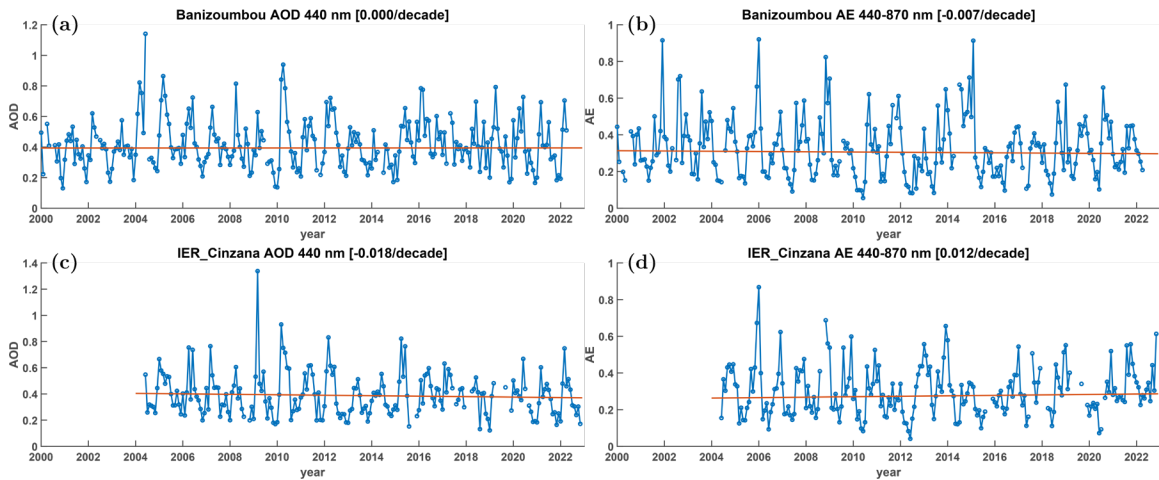


Figure R2: Left: Time series of 440 nm AOD. Right: Time series of 440-870 nm AE. (a, b) Banizoumbou, (c, d) IER_Cinzana.

Technical comments:

- Figure 1: Why is panel (a) labeled as “Solar” Level 2.0? I recommend using the terminology “Direct Sun,” consistent with AERONET products.

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?

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).

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:

"It should be noted that sea salt aerosols typically having FMF_{550} below 0.4 and SSA_{440} 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 statistical significance.

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

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 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.

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 comparison, as explained in the MS in lines 200-202:

"A comparison between AOD₄₄₀ 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 AOD₄₄₀ 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 comparisons in the MS in lines 185-186:

“The rates of AOD₄₄₀ 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, 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 mainly occurred after 2008 (i.e., Osaka, Beijing, and XiangHe). We have also revised the description in lines 194-198:

“However, the trend of AOD₄₄₀ in East Asia is not coherent throughout the period of 2000-2022. According to the AOD₄₄₀ time series (Fig. 5a-c), AOD₄₄₀ 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 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_{440} trend, some other oceanic stations worldwide also exhibit positive AOD_{440} 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.”

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.

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. 5f and revised the trend value to 0.167.

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 Kciclo); can the authors confirm?

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.

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?

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 $AE_{440-870}$ trends, indicating weak changes in the ratio of fine-mode and coarse-mode aerosols. Therefore, the great decrease of aerosol 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 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 the two parameters.

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_{440} trend for Solar_Village (Fig. 13b) in the Arabian Peninsula is attributed to increases in absorbing dust aerosols.”

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.

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

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

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

232 **References**

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1 **Reply to Dr. Collaud Coen**

2 The authors thank Dr. Collaud Coen for her detailed comments and thoughtful suggestions,
3 which are very helpful in improving our manuscript. We have carefully considered all the
4 comments and revised the manuscript and the supplementary accordingly. A point-by-
5 point response to these comments is presented below.

6 **General comments:**

- 7 1. Methodology for trend analysis: The authors correctly chose Mann-Kendall test
8 associated to the Sen's slope, which are both non-parametric methods. The Mann-
9 Kendall (MK) test giving the statistically significance (ss) is however not described
10 in the methodology section. The following points have to be clarified:
 - 11 • MK test has to be applied on serially independent data. This means that MK test
12 without prewhitening can only be applied on time series without ss auto-correlation.
13 In case of ss auto-correlation, prewhitening methods have to be applied. In this
14 study, no mention of auto-correlation is found. I then require from the authors that
15 either to report no ss auto-correlation in all the time series or to use a prewhitening
16 method to minimize the artifacts bounded to serially dependent data. Since the
17 authors cite Collaud Coen et al., ACP, 2020, they should also be aware of the
18 companion paper Collaud Coen et al., AMT, 2020
19 (<https://amt.copernicus.org/articles/13/6945/2020/>) on MK methodology and the
20 associated github repository (<https://github.com/mannkendall>) giving access to a
21 complete MK and Sen's slope routines with prewhitening methods in R, Python
22 and Matlab.
 - 23 • MK test also requires a homogeneous distribution, namely no seasonal cycles. The
24 presence of seasonality in the used time series is clearly visible (e.g. Fig. 4a, b, c,
25 d, f, Fig. 6 b, c, d, e, g and h, Fig. 10 d, e, f, Fig. 12c). Figs. 7, 8 13 and 14 clearly
26 present the trend results for meteorological seasons. The methodology is however
27 not described so that it is not clear if the homogeneity test between season is

performed or not. The paper however describes different trends directions for different seasons (e.g. L 200s). This has to be clarified

- Concerning seasonality, the climate specificities has also to be taken into account. The applied seasons correspond to mid-latitude climate but not e.g. to lands with a monsoon seasonality. The different seasonality has to be taken into account in the analysis.
- As specified in Collaud Coen et al., AMT, 2020, the use of a lower time granularity (e.g. daily) than month could also help to increase MK test's power
- Finally, confidence limits can also be computed and help the interpretation of the results.

Thank you very much for the detailed comments on techniques. We revised the entire trend analysis according to your suggestions, and the details of the method to calculate the significance of MK test has been added in the Sect. 2.3.

The previous results did not undergo pre-whitening or seasonal homogeneity tests. We have attempted to use the algorithm provided by Collaud Coen et al. (2020). We applied the 3PW pre-whitening method and test the homogeneity. However, using monthly data, the majority of stations did not pass the seasonal homogeneity test. As the main purpose of this study is to analyze the multi-year variations of aerosol parameters, we prefer to capture the trends on an annual scale. Therefore, we decide to calculate the annual trend using annual mean data, which have limited auto-correlation and no seasonality. As for seasonal results, we also calculated the seasonal means for each year, and then calculated trends for each season using the seasonal mean time series. Estimating a valid seasonal trend also requires at least 10 years of records. However, the updated results are less likely to show statistical significance due to the reduced sample size.

As for the seasonal analysis, we have introduced additional season divisions for the monsoon (South Asia) and dust source (the Arabian Peninsula and West Africa) regions. Specifically, for South Asia, the seasons were divided into pre-monsoon (March-May), monsoon (June-September), post-monsoon (October-November), and winter (December-February). For the Arabian Peninsula, the seasons were divided into pre-peak (November-

February), peak (March-June), and post-peak (July-October) (Habib et al., 2019). For West Africa, the seasons were classified as Harmattan (November-March) and summer (April-October) (Balarabe et al., 2016; Nwofor et al., 2007). The description about season divisions has been added in the caption of Fig. 8.

Confidence limits are also calculated, and listed in the tables in the supplementary.

2. Homogeneity of the time series

Long-term trend analysis can only be performed on homogeneous time series. The authors reported the case of Birdsville, where false results were reported due to false data filtering. Which procedure was applied to check the homogeneity of the time series ? I do really appreciate to have all time series in supplement. It's worth to have a look if we are interested at one particular station.

Generally, I would really have a look at all time series and remove too high or too low values (e.g. SSA below 0.6), to see if too few data are present in the first or end years so that the time series should be shortened and if there is evident ruptures.

Here some comments on the time series:

- Ames AOD: global decrease but an increase in maxima: to check
- Amsterdam: strange high values in 2010/2011 and 2014
- Anmyon and Arica AOD: is there a rupture due to the long missing period ?
- Bozeman: are high AOD in 2017 and 2021 due to e.g. biomass burning ?
- Cabauw AOD: I would not consider the 2 data in 2003
- Canberra: I would not take the too high 1-3 first data
- Cartel: increasing until 2006 and decreasing after a missing period in 2008-2022: to check
- Ceilap: value > 0.15 in 2012 is doubtful
- Chen-Kung: AOD: I would not use the first two months in 2002, even if MK accept missing data, having a full first and end years remains important. AE: idem
- Davos AOD: I would not take the 2001 Data
- Egbert: do you have an explanation for the high maxima after 2014?
- Fort-McMurry: I would not take 2005 data

- 86 • Hamburg: AOD I would only use 2003-2016 since there is few data otherwise
- 87 • Morin: strange AOD>4 in 2003
- 88 • Issyk : AOD seems very high in 2021
- 89 • Shiraham: I would stop in 2016
- 90 • Osaka: AOD: I would not use the first two months. The maxima are in 2000-2007
- 91 are much higher than thereafter. Is there a change in 2006-2007? AAOD: the very
- 92 high data (> 0.1) should probably be invalidated and the low data end of 2017 to
- 93 mid-2019 are also strange. SSA : similar comment as for AAOD (but inverse
- 94 dependence)
- 95 • Solar village: AOD: seems ok, SSA: the mid 2000-2002 data seems strange and too
- 96 high and max in 2010 as well as min in 2012 should be checked.
- 97 • Gandhi college: the max at 2.5 is very strange and should be analysed. The four last
- 98 months are also much higher after a missing period. Is there a rupture in the time
- 99 series?
- 100 • Carpentras AOD: the first ~6 months are much higher. AAOD: the 2002-2005 data
- 101 seems too high and a rupture in the time series in the missing period (2005-2006)
- 102 is probable. SSA: the three high values in 2006 should be checked
- 103 • Mexico city: AOD: the three low data in jan-feb 2010 are strange.
- 104 • Missoula AE: I would not use the data before 2004
- 105 • Beijing: AAOD and SSA: I would not use the first isolated 2-3 months
- 106 • GSFC: AAOD: the low minima in 2010 and 2011 should perhaps be investigated
- 107 • MD Science Center SSA: the first high value until 2002 and the high values in 2016-
- 108 2028 should be checked as well as the very low values in 2019.
- 109 • Concerning the AAOD, I just looked at some station: Lille has too few data before
- 110 2007, Rome data are really too low in 2012, there is a problem, White Sands: the
- 111 increase after 2019 is so rapid that it is doubtful
- 112 • Concerning SSA. I have the impression that SSA time series are the more uncertain.
- 113 For example, SSA at Granada is not homogeneous at all, 2012-2026 are too low.
- 114 There si various station with very low SSA (e.g. 0.5 at IMAA-Potenza, 0.3 at OHP)
- 115 that should be removed before the trend analysis. I also have the impression that

116 there is rupture in some dataset such as Tucson, Trelew (in 2017?), Toulon in mid-
117 2006, Palencia in 2007-2008.

118 Please have a look at all time series to improve their relevance for long-term trend analysis.

119 Thanks for these very detailed comments and careful observation. We have checked these
120 time series and changed the data filtering strategy. In the previous results using monthly
121 data to calculate trends, the filtering strategy were used solely to select stations with
122 extensive records. For the selected sites, all monthly data were used to calculate the trends,
123 even for years that did not meet the “at least 8 monthly measurements” criterion. Therefore,
124 some years may have data for only a few months, which led to discontinuity of time series,
125 such as Cabauw, Chen-Kung, Osaka, etc., as mentioned in the comment. In the updated
126 results, we switched to use the annual mean data to estimate the annual trends, and data
127 from the years with less than 8 monthly data have been excluded. Only those years with at
128 least 8 monthly data were retained to calculate annual and seasonal means. Consequently,
129 the updated results significantly improve the continuity and homogeneity of the data. Time
130 series of the following stations mentioned in the comment concerning discontinuity data
131 have been greatly improved: Cabauw, Chen-Kung, Davos, Fort-McMurry, Hamburg,
132 Shirahama, Mexico-city, Missoula, Beijing, Lille, and Rome.

133 We also removed outliers for the records, thus time series of the following sites having
134 doubtful values have been improved: Canberra, Ceilap, and Ilorin.

135 For SSA time series, we removed the very low values (below 0.7) from the “all-point” data.
136 The very low SSA values often occur alongside low AOD, meaning that these SSA values
137 are more uncertain.

138 The response to comments for some sites:

- 139 • Ames: The AOD trend is not significant using annual mean value to calculate trend
- 140 • Bozeman: This site is located in western North America. This region was reported to
141 have increased forest fires (Eck et al., 2023; Iglesias et al., 2022), thus the high AOD
142 in 2017 and 2021 is likely due to biomass burning.

- Cartel: This site is located in eastern North America. The AOD time series is similar to that of other stations (i.e., CCNY) in the region. Other studies using satellite observations and AERONET measurements also suggested a slight increase in AOD before 2006 (Zhao et al., 2017; Meij et al., 2012).
- Issyk: The very high AOD in 2021 is likely attributed to strong dust storms initialized by Mongolian cyclone (Yu et al., 2023).
- Osaka: The first two months have been removed because this year did not meet the “at least 8 monthly measurements” criterion (denoted as “Issue #1”). AOD increased in 2000-2007 and then reduced after 2008 in East Asia. The time series is similar to that of Li et al. (2014). The very high AAOD in 2011 has been excluded due to “Issue #1”.
- Solar_village: The strange SSA values occur alongside low AOD levels in non-peak seasons when dust is less predominant, thus the SSA is influenced by anthropogenic aerosols. Moreover, low AOD levels also lead to higher SSA uncertainties. The time series is similar to that of Li et al. (2014).
- Gandhi_college: The extreme high AOD in 2011 has been removed. The four last months happen to be the winter of 2022 when AOD is high. These data have been excluded due to “Issue #1”. There is no rupture in the time series using annual mean data.
- Carpentras: The AOD and AAOD time series are similar to those of Li et al. (2014). The three high SSA values in 2006 has been excluded due to “Issue #1”.

Time series of other sites were also checked, and the values appear to be reasonable.

3. Results reported in a map:

The representation in a map is very useful to have an overview of the trends around the world. I have however some remarks:

- the very small trends (e.g. with AOD slopes in $[-0.02, 0.02]$ (Fig. 3)) are in white but still sometimes ss. Since no table with all results are given, it's not easy to know if the trend are positive or negative. Moreover it means that not ss trend does not appears on the map since there is no dark circle.

- The presented results for all parameters does not correspond neither to the same time period nor to the same length (e.g. AOD at GSFC corresponds to the 23 y trend ending in mid-2022, whereas result from Ghandi-College correspond to 17 y trend ending in 2021 and result from Solar in 13 y results ending in 2013) (+ Fig. 1). My opinion is that trends with up to 10 y differences for the end point or with large differences in the length of the time period should not be represented in a similar way in the same figure. For example, the high positive AOD trend for Solar Village cannot be compare with the Ghandi or Kampur trends since there is almost one decade difference of the end time.

Thanks for the suggestion. We have edited the colorbar to avoid near-white colors. Moreover, the marks of insignificant trends have been changed to triangles with black boundaries. We have also added several tables in the supplementary to list the trends of parameters of all the stations.

Time series of different sites may cover different time periods and have different length, therefore it is hard to sort them into a few categories. Moreover, the maps in the MS are mainly used to provide an overview of the spatial patterns about the trends. Detailed information, such as time periods, could be observed according to time series of individual sites included in the supplementary material.

4. Results with low AOD value and consequently larger uncertainties:

As well explained in the manuscript, low AOD values leads to high uncertainties for the derived parameters. I think that the trends with high uncertainties should appears differently in the map. I don't know what is the best solution. Perhaps by representing only trends with 95% confidence level and different size as a function of the uncertainty ?

Thanks for the suggestion. It is difficult to represent the uncertainties quantitatively. The uncertainties of AE, AAOD, and SSA are all correlated with AOD levels, but they do not have clear relationships. Rough relationships between these parameters and AOD are discussed in lines 127-128 of the revised manuscript:

“According to Eq. (1), the uncertainty of AE is roughly inversely proportional to AOD, with larger errors at lower AOD conditions.”

and in lines 140-141:

AERONET SSA have an error of ± 0.03 when $AOD_{440} \sim 0.4$, and the error increases rapidly (exponentially) at lower AOD levels.”

All of these parameters (AE, AAOD, SSA) have higher uncertainties at lower AOD levels, thus AOD levels could be an identifier for uncertainties qualitatively. We have added a map of AOD in Fig. 3, and added the description about the uncertainties in lines 113-115:

“The patterns of AOD (Fig. 3) and AOD trends (Fig. 4) should be always kept in mind when analyzing trends of the other aerosol parameters, because uncertainties of the other parameters are closely related to AOD level (see below), whose trend reflect changes of aerosol loading.”

5. Data used

It is not easy to understand which data are used. AERONET Solar Level 2 and AERONET almucantar Level 1.5 data are both used, the 1.5 ones for the inversion products. L. 87-88 says that L 1.5 are similar to L 2.0 but for the AOD threshold ? meaning that no AOD threshold are used ? It would be very helpful to have a more precise description with eventually the mention of the level in the figures' captions.

We are sorry for the confusion. This description is generally right. AERONET Solar Level 2.0 data are used in AOD and AE analysis, whereas quality-controlled inversion Level 1.5 data are used in AAOD and SSA analysis. The quality control for Level 1.5 data that we adopt is the same as that for Level 2.0 except the AOD threshold, as explained below.

The reason for not directly using Level 2.0 inversion data (quality assured) is the lack of data samples (fewer than 10 stations), which is caused by the AOD threshold criterion. This has been mentioned in the MS in lines 81-83:

“However, as Level 2.0 quality assurance for inversion products requires a coincident AOD exceeding 0.4 at 440 nm, many stations do not have enough data samples to produce a long-term record.”

Nonetheless, Level 1.5 products have larger uncertainties, which is not suitable to be directly used. As a compromise between data quality and data availability, we apply most

of the Level 2.0 quality control criteria on the Level 1.5 inversion data for smaller uncertainty, only excluding AOD threshold criterion which is an important reason for data loss. Therefore, the amount of data samples is greatly increased.

Minor comments:

1. Are all the average done with median? Are first daily medians computed and then monthly medians or is the monthly medians computed from hourly data ?

Only monthly data is calculated with median. Annual data and seasonal data are calculated from the monthly medians. We have added the description in the MS in lines 93-95:

“For the years with at least 8 monthly measurements, the monthly medians are then averaged to annual and seasonal means, which are used to calculate annual and seasonal trends.”

The monthly medians are directly computed from AERONET all-point measurements. The all-point data has original temporal resolution, which is calculated from every independent observation of direct solar radiation or diffuse sky radiance.

2. L1: there is changes in aerosol composition but also in their concentration.

Thanks for reminding. We have revised the description in line 1:

“Over the past two decades, remarkable changes in aerosol concentrations and compositions have been observed worldwide...”

3. L 10: I would specify that AE correspond to the wavelength dependence of AOD, since AAOD and SSA also depend on the wavelength.

We have revised AE to “AE (computed from the AOD within the range of 440-870 nm)” in line 10.

4. L17-19: long sentence, please rephrase.

We have revised the expression in lines 18-20:

“The reductions of aerosols in eastern North America mainly result from non-absorbing species. Reductions of both fine-mode absorbing species and non-absorbing aerosols are

254 *found over Europe and East Asia, but the reduction of absorbing species is stronger than*
255 *that of non-absorbing species.”*

256 5. L 34: “which mainly located in ...”: please check the language

257 Thanks for reminding. We have revised the description in line 35:

258 “... which are mainly located in Europe and North America”

259 6. L35: It is not possible to consider SSA as representative of the scattering. Please
260 rephrase

261 Thanks for reminding. We have revised the description in lines 35-36:

262 “... and revealed increased scattering aerosol fraction (represented by single scattering
263 albedo, SSA)”

264 7. L84-85: Considerations on the uncertainties of the various parameters are explained
265 at various places in the manuscript. Please sample them at the same place so that
266 the reader can have a direct overview.

267 Thanks for the suggestion. We have revised the MS and moved the description of these
268 parameters as well as their uncertainties in Sect. 2.2.

269 8. L 100 and Figs 1 and 2: Figs 1a and b could perhaps be merged with different color
270 for Level 2 and 1.5? A map (perhaps divided into continents) with all stations’ name
271 could appears in the supplement and/or a table with the stations’ coordinates.

272 Thanks for the suggestion. We have revised to use different colors for Level 2.0 solar and
273 Level 1.5 inversion measurements in Figs. 1a and 1b respectively.

274 We have also added several tables in the supplementary to list the name, location, trend of
275 parameters of all the stations.

276 9. L102: does the AE corresponds to a fit including all the wavelengths between 440
277 and 870 nm?

278 The AE parameter is also a product of AERONET sun direct measurement, and is
279 calculated from the linear regression of AOD and wavelengths on a logarithmic scale

within the range of 440-870 nm (Eck et al., 1999; Giles et al., 2019). All the AOD measurements within the 440-870 nm are used to calculate AE (Giles et al., 2019). This has also been mentioned in the MS in lines 107-108:

“The AE is calculated from all AOD measurements within the 440–870 nm wavelength range (typically including 440, 500, 675, and 870 nm)”

10. Eck 1999

“Eck et al., 1999” refers to the following research article which studied the wavelength dependence of AOD:

Eck, T. F., Holben, B. N., Reid, J. S., Dubovik, O., Smirnov, A., O'Neill, N. T., et al. (1999). Wavelength dependence of the optical depth of biomass burning, urban, and desert dust aerosols. *Journal of Geophysical Research: Atmospheres*, 104(D24), 31333–31349. <https://doi.org/10.1029/1999jd900923>

We have cited this reference in several places in the manuscript.

11. L 123: what do you mean by “all-point”?

We are sorry for the confusion. The meaning of all-point data is detailed in Minor Comment #1. The “all-point” data is a series of AERONET products with original temporal resolution. Detailed information could be found from the AERONET website, <https://aeronet.gsfc.nasa.gov/>.

12. Table 1 and L 121: Why Uncertain is not called sea salt ?

We are sorry for the confusion. We directly applied the names of the aerosol type from Lee et al. (2010), which named aerosols with FMF_{550} below 0.4 and SSA_{440} higher than 0.95 “Uncertain” type. The 0.95 SSA_{440} threshold is mainly used to identify “Dust” aerosols, whose SSA_{440} is typically 0.92-0.93 (Lee et al., 2010). Although sea salt is the coarse-mode scattering species, the SSA_{440} for sea salt is typically 0.98 (Lee et al., 2010). Therefore, the “Uncertain” type includes sea salt aerosols, but not all the “Uncertain” aerosols are sea salt. As “Uncertain” aerosols only take a negligible proportion (2.5%), we

306 did not further classify them into sea salt and a transitional type. We have revised the
307 description about sea salt and “Uncertain” type in lines 170-173:

308 *“It should be noted that sea salt aerosols typically having FMF_{550} below 0.4 and SSA_{440}*
309 *around 0.98 (included in the "Uncertain" type in Table 1) are not considered in the analysis*
310 *of aerosol type trends (Sect. 3.3), because most AERONET stations are located over land*
311 *where sea salt is not the predominant type, and sea salt aerosols only account for a*
312 *negligible proportion (about 2.5% for "Uncertain" type).”*

313 13. L125: it means that the trend results for the various aerosol types are computed
314 from time series with three time less data points due to the seasonal median? How
315 is the seasons defined for monsoon climate ?

316 We are sorry for the confusion. In the updated results, we also used annual mean AOD for
317 each type to calculate trends. We have revised the description in lines 175-176:

318 *“For each aerosol type, we use coincident Level 2.0 AOD_{440} measurements to calculate*
319 *the annual AOD and analyze its trend.”*

320 The seasons (MAM, JJA, SON, and DJF) are defined mainly for the mid-latitude, where
321 most AERONET stations are located. As mentioned in General Comment #1, we have re-
322 defined seasons for monsoon and dust source regions.

323 14. L130-131: This is not the right causality: negative AOD trends demonstrate the
324 global reduction of aerosol loading.

325 Thanks for reminding. We have revised the description in lines 181-182:

326 *“Significant negative AOD_{440} trends are found for the majority of stations all over the*
327 *world, demonstrating a global reduction of aerosol loading.”*

328 15. L135: Higher slope in Li et al. 2014 can also be due to the shortest time series
329 leading to larger slopes due to a much lower number of data.

330 Thanks for reminding. We also agree that the higher slope in Li et al. (2014) might be
331 attributed to a short data record. However, according to the time series of some European

stations (Fig. 5), we could still find that the reduction of AOD has slowed down in recent years. We have revised the description in lines 185-187:

“The rates of AOD₄₄₀ 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. This is also in line with the AOD₄₄₀ time series at representative European sites (Fig. 5g,h).”

16. L139-140: In this case, it is important to know the length and end year of the time series. Do the larger slopes correspond to the shorter time series ? or to earlier end year ?

Thanks for reminding. The larger slopes indeed correspond to the shorter time series. For East Asia, Chen-Kung_ Univ have only 10 years of annual records, and the AOD trend could reach -0.23 per decade. Osaka has longer AOD records, and the slope is smaller.

However, when comparing with other regions (i.e., Europe and North America), the larger slopes in East Asia do not always correspond to the shorter time series, but correlate with its higher AOD levels. For example, Beijing and XiangHe have longer records, higher AOD levels, and larger trends than Brussels and Barcelona. When reducing the same proportion of AOD, higher AOD levels would lead to larger AOD reductions, thus corresponds to larger slopes. In this case, according to the AOD time series, the most considerable AOD reductions indeed occur in East China.

17. L141-144: please rephrase

We have rephrased the description in lines 194-200:

“However, the trend of AOD₄₄₀ in East Asia is not coherent throughout the period of 2000-2022. According to the AOD₄₄₀ time series (Fig. 5a-c), AOD₄₄₀ 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). This result also explains why Li et al. (2014) found no significant AOD₄₄₀ in East Asia with shorter records, as the increase of AOD₄₄₀ in the early 2000s

offset the reduction after 2008. When applying longer records, the continuous reduction of AOD_{440} after 2008 become dominant.”

18. L 147-148: does both time series have the same end year ?

AOD time series of the two sites have different end years. AOD time series of Beijing covers the period of 2002-2018, whereas that of XiangHe covers the period of 2005-2021. We have revised the description in the MS in lines 202-205:

“Both stations possess Level 2.0 records spanning a period of 17 years. However, the data record for Beijing, starting in 2002 and ending in 2018, reveals an AOD_{440} trend of -0.175 per decade, whereas that for XiangHe, starting in 2005 and ending in 2021, is more recent and exhibits a larger AOD_{440} decrease of -0.201 per decade, emphasizing the later years as a period of most notable AOD_{440} reduction.”

19. L 150 and L161-162. The special case of Birdsville should be reported only once in the paper.

Thanks for the suggestion. We have reorganized the paragraph. Discussion about Birdsville and other sites with weak AOD trends has been moved to the second half of the paragraph in lines 213-216:

“Significant positive AERONET AOD_{440} trends over the other regions, such as Birdsville in Australia, Trelew in South America, and Nauru, an oceanic island station, are generally weaker, with magnitudes typically below 0.03 per decade. As these sites have very low AOD_{440} (typically below 0.1 for monthly values) as well as low AOD_{440} variability, the results in these stations are typically more uncertain.”

20. L159-160: are all these trends ss ?

We are sorry for the confusion. We meant to indicate stations with significant positive AOD trends here. We have clarified this in the MS in line 213:

“Significant positive AERONET AOD_{440} trends over the other regions ...”

21. L176: which time series and seasons are less robust due to low AOD ? A map with AOD values (or seasonal AOD) could perhaps help

387 We have added the map of AOD in Fig. 3, and seasonal AOD maps in the supplementary.
388 The description about uncertainties of analyzed parameters has also been added in Sect.
389 2.2 in lines 113-115:

390 *“The patterns of AOD (Fig. 3) and AOD trends (Fig. 4) should be always kept in mind*
391 *when analyzing trends of the other aerosol parameters, because uncertainties of the other*
392 *parameters are closely related to AOD level (see below), whose trend reflect changes of*
393 *aerosol loading.”*

394 22. L179: From the map I see 2/4 stations in western North America have positive AE
395 trends.

396 We are sorry for the confusion. We have updated the result with annual mean time series
397 (detailed in General Comment #1). In the updated map, 2 stations in western North
398 America have significant positive AE trends.

399 23. L198-199: I have the impression that no ss AE trends is just an indicator of no
400 modification of the size distribution. Is it right ?

401 Yes. The statement of reductions in both fine-mode and coarse-mode aerosols is inferred
402 by both no ss AE trend and ss negative AOD trend. We have revised the expression in lines
403 235-238 for clarity:

404 *“East Asia exhibits no significant $AE_{440-870}$ trends, indicating weak changes in the ratio*
405 *of fine-mode and coarse-mode aerosols. Therefore, the great decrease of aerosol loading in*
406 *East Asia revealed in Fig. 4 might be related to similar reductions in both anthropogenic*
407 *fine-mode aerosols and coarse-mode dust in these areas.”*

408 24. L200-201: As mentioned in the general comments, is the homogeneity between the
409 seasonal trends computed ?

410 As detailed in General comment #1, the majority of stations did not pass the seasonal
411 homogeneity test. As the main purpose of this study is to analyze the multi-year variations
412 of averaged aerosol parameters, we updated the results using annual mean data.

413 25. L204-205: Are AOD higher in spring and lower in winter for all stations in the
414 Northern Hemisphere? Here too a map of AOD for the various seasons could help.

415 Thanks for the suggestion. We have added seasonal AOD maps in the supplementary.

416 26. L 229: please rephrase: AAOD does not characterizes the scattering.

417 Thanks for reminding. We have revised the description in line 134:

418 *“AAOD and SSA together characterize the scattering and absorbing properties of*
419 *aerosols.”*

420 27. L234-239: this should be discussed in the method/data section.

421 Thanks for the suggestion. The discussion about the uncertainties of AAOD and SSA have
422 been moved to Sect. 2.2.

423 28. L244: increases in either the concentration of absorbing aerosol or in the
424 composition (higher imaginary part of the refractive index)

425 Thanks for reminding. Changes in either AE, AAOD, or SSA would indicate changes in
426 aerosol compositions, as they suggest changes in aerosol size distribution or refractive
427 index or both. However, in this work, we simply regard aerosols as a mixture of absorbing
428 and scattering aerosols, and analyze the change of aerosol scattering and absorption
429 properties.

430 The reason for AAOD change should be analyzed together with trends of other parameters,
431 especially the AOD trend, which have been added in Sect. 2.2 in lines 113-115:

432 *“The patterns of AOD (Fig. 3) and AOD trends (Fig. 4) should be always kept in mind*
433 *when analyzing trends of the other aerosol parameters, because uncertainties of the other*
434 *parameters are closely related to AOD level (see below), whose trend reflect changes of*
435 *aerosol loading.”*

436 In this case, Solar_Village have positive AOD trends, thus the increased AOD is likely
437 related to increases in absorbing aerosols.

438 29. L262: absorbing (b missing)

Thanks for reminding. We have revised it in the MS.

30. L271-272: Is there not change in BC or BrC concentrations in middle East ?

Solar_Village exhibits significant positive AOD and AAOD trends, as well as negative AE and SSA trends. This means that Solar_Village might have higher aerosol concentration, smaller FMF, and increased absorbing species. As dust is the predominant aerosol, we could infer increased dust activities according to the trends of these parameters.

Changes in BC or BrC is also possible, but we could not infer this according to the trends of AOD, AE, AAOD, and SSA, especially that the significant negative AE trend suggests decreased fine mode fraction. Aerosol type analysis in Sect. 3.3 also suggests no significant trends are found for fine-mode types. Therefore, whether BC/BrC concentration changes needs further research.

31. L 310: I have the impression that, e.g. SSA and AE in western North America, AOD in India or AAOD in Africa have different seasonal trends (Fig. 14).

We are sorry for the confusion. Some regions indeed have different seasonal trends for some parameters, but seasonal results are generally consistent with annual results at the majority of regions. Here we meant to express this similarity in pattern. We have revised the expression in lines 335-336 for clarity:

“Although some regions, such as North India and western North America, have different seasonal and annual trends, the majority of regions do not exhibit significant seasonality.”

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