

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2022-219-RC1>, 2022
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Comment on acp-2022-219

Anonymous Referee #1

Referee comment on "Long-term variation study of fine-mode particle size and regional characteristics using AERONET data" by Juseon Shin et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-219-RC1>, 2022

General comments:

This paper describes the determination of long-term trends of aerosol optical depths (AODs) at 17 AERONET stations in different regions of the world. The AODs are separated into dust- and pollution related fractions, and the pollution-related AOD is further divided into fractions associated with fine- and coarse mode aerosol components. Like the AODs, the total and fine-mode Angstrom exponents were analysed in terms of possible trends.

The topic is suitable for ACP, even though the main result, a reduction of pollution related AODs at many urban sites during the last 20 years, is not surprising. This work is based on experimental and theoretical work of many research groups contributing to AERONET. Using AERONET data for innovative research by other groups is of course in order and welcome but should meet certain standards to value the AERONET project. In my view the current paper is not suitable for publication and does not meet these standards. The numerical data analysis is questionable, formulas and results are partly inconsistent. Moreover, some important aspects are not considered, e.g. whether or not yearly averages of the various variables are suitable to represent conditions at a site (considering the typical high variability of aerosol loads, compositions, data gaps, seasonal effects). Instead, the yearly averages are treated like ordinary measurement values that, apart from a linear trend, scatter statistically. An analysis of uncertainties is missing completely. The years 2019, 2020 and 2021 were excluded from the analysis. It would have been interesting to see how significant short-term effects of lockdown measures were compared to any long-term trends.

Specific comments:

Title: The title is misleading. Fine-mode particle size is studied here rather indirectly. The title should reflect what was done in the study.

Line 28: " τ decreased in Europe and Asian regions and increased in the Middle East, India, and North Africa." After reading the paper, it turns out that these statements are partly incorrect by the authors' own standards of significance. τ decreased significantly in 2/3 of stations in Europe and 2/6 stations in Asia, it increased at 1/2 stations in India and at none in the Middle East and Africa.

Line 31: The abstract should be comprehensible without the remainder of the text. The meaning of "Z-values for Seoul and Osaka are -2.95 and -2.31, respectively" is unclear, a three-digit precision of these parameters is misleading.

Line 32: "...reduction of primary emissions from plants and other anthropogenic sources..."
Unclear.

Line 33: "Values of α_T decreased by -3.3 to -30.5% in Europe, North Africa, and the Middle East, which means" These decreases are all classified as not-significant (Tab. 4). Why is this highlighted as a result in the abstract? Again, the number of digits pretends a high precision that is not justified.

Line 35: "We find that α_T increased by 1.3 to 13.1%." Same problem again. The increase is significant only at 2/8 stations in India and Asia.

Line 35: "In particular, α_{PF} increased in most areas." α_{PF} significantly increased at merely 5/17 stations.

Line 72: "We also analyzed (Dubovik et al., 2002,....)" The use of the citations is unclear. What was the aim of this study, what is new and what was done before and requires a citation?

Line 79: "Second, the depolarization ratio is essential to calculate dust ratio, so we have limitations in using data in American and European sites." This statement is unclear. What are these limitations? And what is different at the three European sites that were selected. What excludes all American sites?

Line 81: "Third, the selected observation sites need to be representative of regional characteristics" Unclear. What are the criteria? Why are, for example, three sites directly at the Mediterranean representative for Europe? Are urban sites representative at all for a region?

Line 107, Tab. S1: The number of observation days is quite small, e.g. around 30 or less per year for the European sites. A quick check for Thessaloniki shows many more days marked as measurement days. What is limiting the observation days in this study?

Line 112: "...we divided aerosol types as dust, fine- and coarse-mode particles depending on depolarization ratio and particle size data". The definition of fine- and coarse mode should be clarified here. According to the AERONET documentary there is a flexible limit in a range between 0.44 μm and 0.99 μm that is determined separately for each distribution. Does this affect the current study?

Line 131: Remove "of particles". This is not the fraction of particles but the fraction of AOD. Is the CMF at 1020 nm an AERONET product? On the webpage I only see FMF (1-CMF) for 500 nm.

Line 132: "Thus, R_D represents the proportion of AOD for pure dust particles in a mixed aerosol plume". If Eq. 3 is correct, this statement is incorrect. The dust ratio apparently describes the dust contribution to (lidar) backscatter coefficient, not AOD.

Line 140: "CMF is higher than R_D in most cases, which implies that these coarse-mode particles include dust and pollution particles". Again, if Eq. (3) is correct, the lidar ratios must be applied here too for direct comparison.

Line 143: "The ratio of coarse-mode particles (CMP) denotes the proportion (number concentration) of coarse-mode pollution particles to total particles". So far, the discussion was on AOD and contributions to AOD. Now it's number concentrations?

Eq. (2): I think the lidar ratios must be applied here too. Moreover, the underlying assumption is that all dust particles belong to the coarse mode fraction. Therefore, CMP will be a lower limit and the FMF (pollution) an upper limit because the fine mode is then considered pollution only?

Eq. (3): The lidar ratios were not considered before, as already mentioned. I think the equation requires an explanation and/or citation.

Line 150: "The parameter S is the lidar ratio of the aerosol mixture. S can be calculated from the AERONET data products." Did you calculate this ratio, or is it part of the AERONET data products? It would be interesting to know how strongly the S can deviate from the S_d .

Line 154: "The AOD of coarse-mode particles (τ_{PC})..." Shouldn't that be coarse-mode **pollution** particles?

Line 158: "The term a_{PC} is the Ångström exponent of coarse-mode pollution. We used the value of 0.16 and 0.14 for Asian and Saharan dust, respectively (Shin et al., 2018)." Why do you use a dust-value for coarse-mode pollution particles? The Angstrom exponent is not only a function of size but also of aerosol type. I would expect much greater values.

Eq. (6): $\tau_{D,\lambda}$ is probably calculated as in Eq. (5)? Anyway, this equation needs more explanation. I understand the approach if you would only have CMF_{1020} . But as far as I can see AERONET provides FMF_{500} and Angstrom exponents for the fine-mode and the total AOD. So there seem to be other possibilities or options to check your calculated $\tau_{PF,\lambda}$. Please clarify which AERONET quantities were used and which quantities were derived from these in detail.

Line 165/166: What is the difference between "First,..." and "Second,..."?

Eq. (7)/line 170: N should be the time span in years rather than the "number of available data" because in some of your time records there are gaps. If you define the percentage change this way the data in the tables are not comparable.

Line 172: "The p -value (probability value) is a scalar that describes how likely it is that the data occurred by random chance. The p -value should be small enough, depending on the level of confidence." How were the p -values calculated and how "small" should they be?

Line 172: "We calculated all data and found a statistically significant trend". Certainly not for all sites and quantities.

Line 177: "The magnitude of the trend can be explained by Sen's slope test, which is a powerful method for investigating linear trends." What was calculated and what does it mean? The "powerful method" is not used at all in the following.

Line 178: How were the Z -values calculated? As for the p -values and the S , the details should be given somewhere, at least in the Supplement (if you really need them all). Otherwise, no one can reproduce these numbers.

Line 197: "...based on a threshold value, denoted as R_D , which..." R_D was introduced before as dust ratio not as a threshold.

Line 200: "In this study, we used seven aerosol types based on the values based on the values of δ_p , respectively." It's unclear if a new classification is introduced here or if the method by Shin et al., 2019 was adopted. What means "..., respectively"?

Line 205: "The results of our aerosol classification reflect the characteristics of the six regions well." What does "well" imply? Well, in terms what is expected anyway? Well, in terms that the stations are representative for a region?

Line 241: "...we compared trends in the average values of τ_T for each region and site during 2001–2018 (supplementary Figure S2). The average value of τ_T at 440 nm was highest (1.22 ± 0.76) in Beijing..." Fig. S2 clearly shows annual averages but these data do not fit to the results in Tab. 1. For example, the values for Beijing range from about 1.05 to 1.45 in the figure while in Tab. 1 a mean of 1.22 is listed (reasonable) with a standard deviation of 0.76 (unexplained).

Table 1: A look at Tab. 1 shows that the simple budget equation (6) is not fulfilled, especially at the sites with high dust contributions, i.e. $\tau_D + \tau_{PC} + \tau_{PF} > \tau_T$ I wonder if this has to do with missing lidar factors in Eq. 2 or problems with Eq. 5. Moreover, I cannot reproduce the FMF from $FMF = \tau_{PF}/\tau_T$ or $FMF = \tau_{PF}/(\tau_D + \tau_{PC} + \tau_{PF})$. What does all that mean for the trend analysis?

Line 247: "We find an average value of $-0.0138 \tau_T \delta \square \square \square \delta \square \square \square^{-1}$ " This rate of change corresponds to the slope of the red dashed line in Fig. S2 (f). From that a total change of 0.22 in 16 years can be calculated. Divided by the mean of 1.22 gives an -18% change according to Eq. 7. However, in Tab. 2 a change of -33% is listed.

Line 249: "The Chiang Mai and Bangkok sites in Southeast Asia showed a decrease of τ_T , too." This statement is not justified. Looking at Fig. S2 (e) there is clearly no trend at both sites. For Chiang Mai this is confirmed by the small Z-value in Tab. 3 while for Bangkok a Z-value of about 2 is reported which indicates a significant (negative) trend. The slope listed in Tab. 2 is merely -0.0012, hardly visible in the figure and certainly not significant given the scatter of the yearly data.

Table 2: The percent variations given in brackets are not very helpful. First, they are not comparable because they refer to different time spans for different sites. Second large numbers do not automatically mean that the trends are significant.

In Figs. 3 and 4 slopes corresponding to significant trends could be colour coded. This would give a quick overview without having to consult Tab. 3 and 4 for each site.

In Figs. S2-S7 standard deviations of the yearly averages should be added. I assume this will reveal how variable the different quantities really are. Something that is not discussed in the trend analysis.

In Table 3 and 4 dutifully all statistical results are listed but what do they mean? Are the results using Z , p and S as indicators consistent? Can anything be learned from using the different approaches? What does a dash mean? Why do the Z -values have no signs consistently? Moreover, significant digits should be considered. If your criterion for significance is whether Z is greater or lower than 1.7, why report 2.3062? Some of the Z -values are numerically identical which indicates what a crude number that really is for a limited number of data points.

Line 250-383: The remainder of this section is very confusing because of the great number of sites, slopes and potential trends discussed. It is difficult to see what is relevant here and what not. I didn't check everything in detail. The authors must make sure their statements are correct and in fact backed by the data in figures and tables. I suggest to only discuss those trends that were found significant. Example from line 297: "... but the percent variation was as high as 112.29% (Thessaloniki)". This appears to be a large, high precision increase but it's not significant (Tab. 3).

Technical corrections

Line 36: "... showing the probability that ..." Rephrase.

Eq. (4) should probably read $\tau_{PC,1020} = \tau_{1020} \times CMP_{1020}$

Line 121: "...show a comparably high correlation". What is comparably high?

Line 133: Remove "particles" and "particle" to clarify.

Line 150: "Here, 1020 denotes the wavelength at 1020 nm." That should be clear enough.

Line 160: "equation (5)". Should read "equation (6)", I assume.

Line 166/167 and 169/170: You should consistently assign slope and intercept with a and b .

Line 192: "Results" and Discussion