Comment on acp-2021-1012
Anonymous Referee #2

Referee comment on "Airborne observation during KORUS-AQ show aerosol optical depth are more spatially self-consistent than aerosol intensive properties" by Samuel E. LeBlanc et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1012-RC2, 2022

Review for Atmospheric Chemistry and Physics

Title: Airborne observation during KORUS-AQ show aerosol optical depth are more spatially self-consistent than aerosol intensive properties


General Comments: The authors present a method, based on autocorrelation, to examine the spatial and temporal variability of AOD, AE and FMF over South Korea and adjacent waters during the KORUS field campaign which occurred in May-June 2016. There is much interesting data and analysis to digest in this paper, however there are also some key missing aspects that need further discussion and are very important regarding the conclusions reached.

Some discussion regarding the relative uncertainty of the AOD and AE needs to be added to this manuscript. The AE parameter in general has significant uncertainty due to the individual uncertainties of the spectral AOD that are utilized as input to the calculation. See Kato et al. (2000; JGR) equation 6 and Hamonou et al. (1999; JGR) equations 1-3 for estimates of the uncertainty of AE computations. Also note that the uncertainty of AE increases as AOD decreases while the uncertainty of AOD remains constant for all AOD levels for a sunphotometer such as 4STAR. This is quite important in relation to the analysis presented in this paper, and does not seem to have been considered. Further the range of values of measured AOD and computed AE differ significantly (histograms shown...
in Figure 4) with the 4STAR values of AE varying over a relatively small range of values \( \sim 0.7 \) to 1.5. The data sample ranges for these parameters coupled with the greater uncertainty in AE relative to AOD also has significant influence on the statistics computed and compared for these parameters and requires further discussion. For these reasons of expected noisier data for AE versus AOD, I have some doubts that the AOD is more spatially consistent than the AE during this KORUS campaign interval data set, at least to the extent suggested. Also the FMF as computed by the SDA algorithm utilizes as the primary input the AE (at 500 nm) and the spectral derivative of AE which has an even greater uncertainty. Therefore the FMF from SDA also has a significant uncertainty (larger than AOD) that also increases at lower AOD levels. Furthermore, the SDA retrieval assumes bi-modality of the aerosol size distribution while in reality three modes may sometimes exist. Specifically in S. Korea the presence of a middle or third mode (of sub-micron radius) from fog processing of sulphate species is often associated with fog over the northeastern Yellow Sea, as documented in Eck et al. (2020) on some of the KORUS flight days.

Specific comments:

Line 65: Angstrom needs to be capitalized since it is a proper name.

Line 124: This is misleading as the primary data for this study is from May to mid-June (not May-July as you stated), encompassing a time interval of 41 days.

Line 136: It could be argued that the economic booms in eastern China and S. Korea and concurrent increases in industrial pollution may have begun a decade earlier than 2010. Please cite more references and evidence for a dramatic increase in fine particle production since 2010.

Line 185-185: The decrease in transmittance on the optics window for each flight is a significant source of AOD uncertainty. In Table A1 this decrease in transmittance at 650 nm is given for each flight. However it is well known that deposition of film on optics windows results in transmittance decreases that differ with wavelength. This issue should be discussed as it has a greater direct impact on the Angstrom exponent (in comparison to a single wavelength AOD) since multiple wavelengths are used in the computation of AE.

Line 200-201: This is inconsistent with the Section 2.3 title which states Angstrom Exponent is retrieved with GOCI while here in the text you only mention FMF. Which GOCI parameter is actually retrieved and/or used in this section of the paper?
Line 211-212: It would be appropriate and useful for the reader to give the validation/comparison statistics for the GOCI YEAR product of AOD and AE and/or FMF versus AERONET values here in the text.

Line 220: Does MERRA-2 really assimilate the AOD product from MODIS or the clear sky radiances from MODIS which are then converted to AOD by an AI algorithm trained with accurate AERONET measurements of AOD? Please check and clarify.

Line 269-270: It seems that this might be an appropriate place to discuss the flight altitudes of the DC-8 aircraft during this campaign and the lower flight level relative to the faction of AOD below that typical lower flight altitude. At times this paper seems to suggest that total AOD is being investigated but in reality the lowest layers with highest aerosol concentrations are sometimes missing. This is important as much aerosol dynamics (physical and chemical) occurs in the lower boundary layer. This issue needs more discussion/clarification in the text.

Line 301-302: Please note here in the text how many flight days were utilized, plus how many total flight legs.

Line 307-308: Please be more specific of the distance of these shortest autocorrelation bins.

Line 336-337: It should be discussed here that AERONET requires the 380 nm AOD for L2 retrievals from SDA due to the more robust characterization of alpha' (or curvature; derivative of AE) when utilizing this wavelength. It is surprising that the shortest wavelength considered in your computation of SDA was 452 nm. Also, AERONET uses 5 channels (380, 440, 500, 675, 870 nm) as input to SDA retrievals, not 4 channels as stated here in your text.

Line 354, Section 4.1: It would be appropriate to make the same type of analysis for AE including maps such as shown in Figure 2 but for AE instead of AOD. Then the spatial variance of the two parameters could be examined at this spatial resolution.

Line 390: What were the minimum, average and median altitudes flown for these transects analyzed in this paper.

Line 392-393: The number of days sampled in each grid box would be an important statistic to show since some boxes seem to have small sample size and therefore have a non-representative number of days sampled.
The AOD spectra in Fig 3b look somewhat noisy with local minima from ~600 to 625 nm which is also the Chappuis ozone maximum absorption region. I am surprised the AOD spectra do not look smoother than this in logarithmic coordinates. This has implications for the accuracy of the computed Angstrom Exponent and needs to be discussed in the text.

Twenty sampling dates although large for an aircraft campaign is not statistically a very large sample size. Plus I suspect there are many fewer days in some parts of the KORUS domain shown in Figure 2.

This is clearly an exaggeration to say that 5.8% of the variance explained is less than 6.6% of the explained variance, in a statistical sense. They are essentially equal for all practical purposes, within less than 1%.

This is significant, especially if the level flight legs miss a significant portion of the total column AOD due to restrictions on the minimum flight altitude. The portion of the total column AOD that are missed by the lowest flight legs needs to be estimated and documented in the text of this manuscript.

This is also important, as the upper layer has a higher coarse mode fraction and is likely more homogeneous in AOD due to mixing and dispersion in time and space from distant dust source regions.

The AE is lower below 2 km during the extreme pollution/transport regime due to larger size fine mode particles for those dates, see Table 1 in Eck et al. 2020 for the large fine mode radius during this time period. Therefore I think you are mistaken to identify this as greater coarse mode influence due to lower AE. The FMF from SDA should still be quite high for this extreme pollution/transport period. This highlights an issue with the AE parameter (as computed from the wavelengths you used) since it is not always indicative of fine/coarse mode relative influence. The aerosol during the extreme pollution/transport period is affected by humidification plus fog and/or cloud processing during this high cloud fraction and high humidity time period.

Fine mode aerosols are never subjected to 'only transport' as you suggest here, since aging processes such as coagulation occur, and condensation occur during transport plus possible interaction with clouds/fog and particle humidification. The fine mode dominated in all but one flight day.

I disagree that this would only occur within a small distance. After long distance transport fine mode properties can be significantly modified by cloud/fog processing within droplets with subsequent droplet evaporation yielding different particle properties.
Line 575-576: What is the average flight altitude of these data in Figure 9? Please give the mean, median, minimum and maximum altitude as this is pertinent to the informed evaluation of these results.

Line 593: This is not really total column AOD from 4STAR. How much of the AOD is below the flight altitude it unclear as this has not been adequately discussed in the manuscript.

Line 684-685: Please state how many days of 4STAR data were utilized in each of the KORUS meteorological periods. I suspect that the sample size in terms of number of days is not very robust in most of these periods.

Line 712-715: There was also some sea breeze pushing back and forth of aerosol over from the Yellow Sea to over land (and vice versa) during the stagnation period, not just local effects of aerosol production and evolution. Additionally, this period had the lowest AOD and therefore the largest uncertainty in AE and FMF. In fact the uncertainty in AOD approaches the AOD magnitude in the long wavelength visible and NIR during the stagnation period.

Line 734-735: There was no evidence of significant dust transport during the extreme pollution transport period as you seem to imply here. The total column FMF is very high from AERONET data using SDA for this meteorological period. Please provide evidence of this dust if you have it since no other published KORUS paper had documented that phenomenon for this particular meteorological period.

Line 735-737: Also it should be noted that this transport period had the highest AOD and therefore the smallest uncertainties in both the AE and FMF. This period has very large fine mode particles related to the high cloud fractions and high RH therefore strongly suggesting particle humidification and/or cloud/fog droplet processing.

Line 737-738: Note that there is evidence for new particle formation in the extreme pollution transport period. See the increased PM2.5 in central Seoul versus the west coast during this interval (Eck et al., 2020). However your flight lines may not be able to identify this phenomenon as it is manifested in surface PM, possibly not at the flight altitudes of the flight segments which were analyzed in this study.

Line 762: Please give the number of days of sampling for each of the three altitude layers.

Line 832-833: This is an odd emphasis in the Conclusions section on dust at higher
altitudes as the total column AOD during the extreme pollution period was dominated by fine mode aerosols.