Review of manuscript acp-2020-1250

GENERAL REMARKS

The article by Titos and co-workers presents a data rich and extensive analysis of aerosol light scattering properties at elevated relative humidity, based on observations from various sites on the globe. Covered aerosol types reach from polluted urban aerosol to aerosol at remote Arctic and mountain stations. Thus, the study presents an excellent overview of the influence of aerosol hygroscopic growth on the aerosol optical properties. One major target of the study was the attempt to deduce correlations between hygroscopic growth and resulting light scattering properties at ambient conditions and other aerosol properties which are measured more frequently than the more difficult-to-access hygroscopic growth behaviour. Although the study cannot recommend clear parameterisations of aerosol hygroscopic behaviour and their impact on the aerosol optical properties, it makes a substantial contribution to the highly relevant research question of the adequate representation of aerosol optical properties at ambient conditions in global models for the full variety of atmospheric aerosol types.

The paper is very well written and structured, and the number and quality of figures is adequate, with the exceptions of Figures 1 and 6 for which small changes are recommended. The study fits well into the scope of the journal and will be suitable for publication once few issues as discussed in the following have been considered.

SPECIFIC COMMENT

Some of the paragraphs in the Results and Discussions section are very descriptive. Adding a more in-depth physical interpretation of the observations would benefit the manuscript very much. Since these changes may require some work, the requested revisions are rated as Major.

1. Section 3.1 and description of Figure 1: The figure itself presents a huge data set but drawing conclusions from the present version is difficult. The authors state that the datasets from the different sites are not comparable since the sizes of the data sets and the covered seasons may differ significantly from site to site. Although it is almost impossible to present this large and diverse data set consistently, some improvements may be possible. One is to add the sizes of the datasets to Figure 1 (see also Minor Issue 6 below). In the description of the datasets the scatter of data is partially explained by the fact that, e.g., the marine sites may contain polluted as well as clean episodes, but how are “polluted” and clean” defined and why not separating the “polluted” from the “clean” air masses? By combining all datasets from one site to a single analysis, natural variability may cover correlations between air mass characteristics and aerosol hygroscopic growth. Furthermore, the explanation of Figure 1 is very descriptive without discussing physical properties (typical size distributions, chemical composition) which may explain the observed growth factors and may explain some of the observed agreements and disagreements between observations at different sites.

2. On line 325, it is mentioned that for rural stations there is no difference in hygroscopic growth observed between the size cuts of PM_1 and PM_{10}. However, looking at the bottom left panel of Figure 3, the dependence of $f(RH=85\%)_{PM1} – f(RH=85\%)_{PM10}$ shows an almost similar behaviour as for the other aerosol types, i.e., larger differences at smaller values of the scattering Ångström exponent which indicates enhanced presence of larger particles, and decreasing differences with increasing

scattering Ångström exponent values, which indicate the presence of a strong mode of small particles. It is recommended to revisit the discussion of Figure 3.

3. Likewise, the discussion of Figure 4 should be reworked. It is an interesting way of plotting the differences in humidity growth factors. As described in Section 2.1, f(RH, λ) values were determined from measurements of aerosol scattering coefficients with dry and humidified Integrating Nephelometers, using Equation (3). If this procedure is correctly understood, then the measurements include intrinsic size distribution information, since the particle size distribution has a direct impact on the aerosol scattering coefficient. It would be very helpful to see a discussion of physical properties in the description of Figure 4 which can explain the observations.

4. Section 3.3 on the Changes in RFE is very well written and explains very clearly the observed behaviour. It may serve as an example for the sections discussed above.

MINOR ISSUES

1. Please check the spelling of the term Ångström throughout the text to ensure consistent use. Sometimes it is written with a capital "A", sometimes with a capital "Å".

2. Equation (4) is misleading since the independent parameter of the Ångström exponents is a wavelength pair and not the difference between two wavelengths. It is recommended to write α_{sp} (λ_1, λ_2) instead of α_{sp} (λ_1 - λ_2).

3. Line 226: Rearrangement of the sentence is recommended for better understanding. Suggested change is "Figure 1 shows the dry total scattering (top plot) and f(RH=85%) statistics (bottom plot) for individual sites …".

4. Line 232 ff: The authors mention their results suggest "that aerosol loading does not control the magnitude of f(RH=85%)". But how should that be the case since hygroscopic growth depends on the composition of the particles and not on their total load? I suggest adding a short explanation here why this dependence is not observed or remove the statement.

5. Line 436: Suggested rephrasing: the authors may write "This is consistent with the hypothesis that ..." instead of "This is consistent with the idea that … ". There are some other sentences in the manuscript, where a more precise language is recommended.

6. Figure 1: it is mentioned in the text that the sizes of the data sets may differ largely between the different sites. Adding the size of the datasets to the individual box and whiskers per site would certainly help interpreting the figure adequately.

7. Figure 6: The numbers of single-scattering albedo (top panel) and scattering Ångström exponent are difficult to read. The same holds for the numbers on the figure legend. Enhancement of numbers is suggested.