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Interactive comment on “Aerosol optical properties in the southeastern United States in summer – Part 1: Hygroscopic growth” by C. A. Brock et al.

Anonymous Referee #2

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Brock et al. use airborne profiles of aerosol properties performed in the southeastern United States to investigate the hygroscopic effect of aerosol particles on their extinction coefficient at three distinct relative humidities (RH). A new parametrisation is presented that is found to more adequately describe the observations than a different single-parameter equation (γ -equation).

Although I have no doubts as to the high quality and scientific significance of the presented data, I see some major shortcomings in the analysis and the following interpretation. First of all, the paper uses airborne profiles of measured optical extinction enhancement factors (plus size distribution and composition measurements) but no

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profiles or interpretation of these are shown (in contrast to the title of the presented manuscript). This could be due to the fact that these will be shown in a companion paper. However, it is not acceptable that the data selection criteria that lead to the presented results are not accessible for the review process (quote from page 25699, line 20: "Part 2 also contains details of the selection of data for the analysis of both papers ...").

Secondly, the proposed parametrization is based on measurements at one dry and two elevated relative humidities only, which is an understandable and necessary compromise for airborne measurements. However, I doubt that three values are sufficient to propose a new and improved parametrization bearing in mind that many aerosol types show hysteresis effects which can not be described by a simple γ -fit. Due to the limited number of elevated RH measurements it is important that the authors present and discuss the (hygroscopic) calibration of their instrument with nebulized salts prior to each campaign or flight.

The paper would be substantially improved by

- (a) more focused discussion of the actual profile measurements of the extinction enhancement, its vertical and spatial variation, its link to chemical composition and air mass origin (as the manuscript title also implies); and
- (b) less focus on the parametrization of a three point measurement.

There are further detailed comments given below (in arbitrary order) that should be adequately addressed before this manuscript can be published in ACP. It is for these reasons that I recommend major revisions.

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Detailed comments:

- Page 25700, line 21 and page 25701, line 6: What is the exact range of measured RH inside the cells? Please give the range and standard deviation. How stable is the RH during a profile? The current values given are very vague. The dry value changes within the manuscript from <15 % to <25 %. At 25% RH the influence of hygroscopic growth of organic substance can become relevant.
- Page 25700: As mentioned above, it should be clearly stated which profiles were selected and which criteria were applied. Showing the average profiles of $f(\text{RH})$ and case studies would improve the quality of the paper.
- Page 25701, line 10: Were the calibration measurements with ammonium sulfate conducted before each campaign or flight? Please show (at least in the reply letter) the corresponding calibration plot and state the degree of agreement in the revised manuscript. Presenting this information is critical to evaluating the usefulness of the proposed parametrization.
- Sect. 2.1: How were particle losses (from the inlet to the CRDS and within the CRDS) characterized and how were they treated in the data analysis?
- Page 25702, line 7: The low amount of particle light extinction above 700 nm is surprising to me. What was the exact range for all analysed profiles? Could this be due to losses inside the aircraft sampling lines? In the reviewers opinion, it is important to keep the influence of the coarse mode in mind since it has (even at low number concentrations) an important impact on the overall $f(\text{RH})$ (see e.g. Zieger et al, 2013, 2014).
- Sect. 2.2: How were the Mie ambiguities (or Mie wiggles) treated in the UHSAS size correction?

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- Sect. 2.3: The usage of κ -Köhler theory to calculate optical enhancement factors and related closure studies has also been done in previous studies, which should be referenced here (see e.g. Zieger et al., 2013).
- Sect. 2.4 (second last paragraph on page 25709): Please state also the relative uncertainties in $f(\text{RH})$ for the three RH channels.
- Result section, Fig. 2: Is this a ground measurement or why is it not shown as a profile? If it is a ground based measurement, why has it been chosen? Again, it would be highly desirable if the actual profiles would be shown and discussed.
- Fig. 2b: How does the actual $f(\text{RH})$ (calculated vs. measured) compare at the three RH's?
- Sect. 3.1: Defining the γ -parametrization at $\text{RH}=0\%$ as 1 cannot be realistic for actual atmospheric conditions (due to hysteresis effects). I understand that this assumption is needed due to the limited amount of RH channels, however, a systematic error in one of the elevated RH cells could also explain why the γ -fit is not a suitable parametrization. This is one reason why the authors have to include their instrument calibration with known hygroscopic salts and a discussion on particle losses in the revised manuscript. In addition, I find it astonishing that only a short time period (8 July 2013, 11:10-11:45) is chosen for their fit (Fig. 4a) and time series (Fig. 2a), from which it is even not clear if it is an actual airborne profile or not.
- Page 25711, line 26: I doubt that the change in refractive index from dry to elevated RH only has an approximate effect of 20% and thus can be ignored to first order. For example, if one takes the hygroscopic growth factor of a 200 nm ammonium sulfate particle (at $\text{RH}=90\%$) to be approx. 1.7 then this will give at $\text{RH}=90\%$ a refractive index of ~ 1.36 (if calculated using a volume mixing ratio).

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The change in scattering alone will be more than 20% (at RH=90%) and depending on the mode diameter up to 1 μm it will be rather a factor of ~ 1.5 to 2.5. Please clarify and proof your assumptions e.g. by showing a further sensitivity study that accounts for changes in refractive index (e.g. by adding the extinction efficiency for $m = 1.36$ and $m = 1.33$, for pure water to Fig. 5).

- Page 25712, line 16: I suggest to delete this sentence since an argument is brought forward in terms of a measurement result that has not been published or reviewed yet and this is hard to evaluate or judge on (and I guess it is not winter 2015 since this lies in the future).
- Page 25715, line 16: It is not clear to the reviewer why the uncertainty of the organic κ should cancel out when calculating $f(\text{RH})$. A slight overestimation of the organic κ will also lead to an overestimation of the $f(\text{RH})$ (see e.g. Fig. 9a in Zieger et al, 2015) and vice versa. In general, due to the importance of the organic contribution the uncertainty of the organic κ will easily compensate other systematic errors. Please clarify.
- Page 25716 (first paragraph) and Fig. 8: Could these low organic κ -values also result in the effect of a temperature dependent evaporation of inorganic components (like ammonium nitrate, see e.g. Aan de Brugh et al., 2012), which were held constant to derive the organic κ ?
- Table 2 is not discussed in the main text. The comparison in terms of linear regression will always be better for the extinction coefficient which is determined mainly by particle number concentration. To better evaluate the model performance to hygroscopicity measurements the authors should add a table or plot showing the comparison of the actual $f(\text{RH})$ modelled vs. measured. In addition, the variation in RH and the contribution of the coarse mode for each profile should be stated. Again, why were only these eight days selected?

- Fig. 3: What temporal averaging was applied here? In general, how was the data averaged?
- Fig. 4b: For completeness, the ratio of calculated to measured $f(\text{RH})$ at $\text{RH}=90\%$ should be shown as well.
- Fig. 7: Are the outliers determined by certain events or aerosol characteristics? Have the authors tested the different possibilities, e.g. by color-coding the points according to time, flight altitude, temperature, apparent coarse mode, mean geometric diameter? This could be a useful addition to evaluate the model performance.
- Sect. 3.4 and Fig. 8: Please specify how the organic mass fraction was calculated (e.g. was BC included or not).

Technical comments:

- Page 25703, line 3: I would remove 'of a design now commercially available' since it is not relevant for this work.
- Page 25700, line 5; page 25712, line 22: Full stop missing at the end of the sentence.
- Page 25715, line 20: Remove the 'r' behind k_{chem} .
- Page 25697, line 2: I would suggest to remove 'meteorological, trace gas, and' since this data is not being used in the following article.
- Table 1: Please mention the corresponding wavelength in the caption and unit for the density.
- Fig. 6: The x-axis is a little bit confusing and at first the reader looks at 2, 3, etc. μm diameter. Better replace it by 0.2, 0.3, etc.

References:

- Aan de Brugh, J. M. J., Henzing, J. S., Schaap, M., Morgan, W. T., van Heerwaarden, C. C., Weijers, E. P., Coe, H., and Krol, M. C.: Modelling the partitioning of ammonium nitrate in the convective boundary layer, *Atmos. Chem. Phys.*, 12, 3005-3023, doi:10.5194/acp-12-3005-2012, 2012.
- Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, *Atmos. Chem. Phys.*, 13, 10609-10631, doi:10.5194/acp-13-10609-2013, 2013.
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- Zieger, P., Aalto, P. P., Aaltonen, V., Äijälä, M., Backman, J., Hong, J., Komppula, M., Krejci, R., Laborde, M., Lampilahti, J., de Leeuw, G., Pfüller, A., Rosati, B., Tesche, M., Tunved, P., Väänänen, R., and Petäjä, T.: Low hygroscopic scattering enhancement of boreal aerosol and the implications for a columnar optical closure study, *Atmos. Chem. Phys.*, 15, 7247-7267, doi:10.5194/acp-15-7247-2015, 2015.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 25695, 2015.

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