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Atmospheric Chemistry and Physics
Discussions

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

C. A. Brock et al.

charles.a.brock@noaa.gov

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The discussion below includes the complete text from the reviewer, along with our responses and corresponding changes made to the revised manuscript. The authors thank the reviewer for useful comments that have improved the manuscript.

We note that the manuscript has been substantially reorganized in response to Reviewer 2. All line and page numbers below refer to the original manuscript. A PDF of the revised manuscript with major changes highlighted in yellow is attached as a supplement to this comment.

Comment: Scientific significance: Good – Hygroscopicity of aerosol is still highly uncertain, despite being an important factor in determining aerosol direct radiative forcing. The data on OA K_chem are therefore of high scientific importance, especially since they fall on the low end of the expected range and are supported by a robust analysis. The new fit parameter to describe hygroscopic growth, Kext, is a moderate improvement over the most commonlyused fit parameter, gamma, and is of sufficient value to merit publication.

Scientific quality: Excellent – This paper combines careful measurements, quantification of uncertainties where this is possible and acknowledgement where it is not, and a robust, comprehensive and clear analysis.

Presentation quality: Excellent – The paper is very well-written and organized. The analysis presented supports the conclusions. I wish all papers I reviewed were this well-written. This paper should be accepted for publication once the following minor points are addressed:

1. The Abstract and Conclusions state that the new Kext parameter formulation does a better job of describing the observed aerosol hygroscopic growth than does the traditionally-used gamma fit parameter/formulation. This is true, but the improvement over the gamma fit is only, on average, 20% for the RH (70%) where the bias in the gamma fit is greatest; at other RHs the gamma fit is better. By not quantifying the improvement in the Abstract/Conclusions, the reader gets the sense that improvement by using the new Kext over the gamma fit is perhaps greater than it is. Please quantify the improvement in the Abstract and Conclusions.

Response: The manuscript has been modified to focus on the observed hygroscopicity rather than the new parameterization. We note that there are much larger potential differences in the two parameterizations for RH > 90%, and show evidence in the Appendix and Supplemental Materials that the κext parameterization does a better job describing hygroscopicity over the full range of RH values.

Comment: 2. As noted in the comment by Anne Jefferson, the goodness of the gamma fit/formulation will depend in part on what is used for RH_o in Eqn. 1, and this should at a minimum be acknowledged in the paper.

Response: We include a plot of the gamma parameterization using RH_0=35 % (Fig. 7a), which indeed produces a better fit to the medium and high RH values. But this approach is unphysical for a constantly deliquescing aerosol, which we believe is the case for the organic-dominated composition in the southeastern U.S. Additional data presented in the Appendix and Supplemental Materials (for different environments than we measured here) also show a more continuous deliquescence curve for the majority of cases. And lacking additional information, what value of RH_0 should we choose? This essentially makes the gamma parameterization a two-parameter fit, with gamma and RH_0 as the fitted variables. We strongly prefer to use a physically based parameterization that approaches reasonable values at the lower limit of atmospheric RH conditions, and that better simulates f(RH) for RH >90% as shown in the Appendix and Supplemental Materials.

Comment: 3. pg 25705 lines 5-12: "The parameter Kchem may be calculated from the volume weighted contribution due to species i, Ki, which are determined: : :" Please be explicit here: contribution to what? contribution to hygroscopicity? to mass?

Response: This sentence is rephrased to: "The value of κchem for the mixed particle composition may be calculated from the volume weighted average of the κchem of each species i, κi, that contributes to the aerosol composition."

Comment: 4. pg 25707, line 21: Sub-micron sea salt was assumed to be zero. Is there any evidence to support that sub-micron sea salt was negligible? Given its high f(RH), even a small mass contribution might significantly affect extinction at higher RH.

Response: We did not measure refractory aerosol composition on the aircraft. Contemporaneous measurements (Guo et al., 2015) at the SOAS surface site in Centreville, Alabama (near which we flew several profiles and low passes for purposes of com-

parison), show a negligible fine mode contribution from sea-salt. This is now stated in section 2.3.

Comment: 5. pg 25709, lines 24-27 & Figure 2 & Figure 4: Figures 2 and 4 only show/include data (and regression) for a section of one flight. a) Why only fit data from 11:10-11:45 from that flight? b) What is the fit/regression for the full data set? c) How is the comparison of the Kext and gamma fit affected by inclusion of more of the field data? d) Fig. 2b. There seems to be two groups of data: <50 Mm-1 there is excellent agreement; >50 Mm-1 the calculated extinction is higher than the measured extinction. This is clearly the case for another high-extinction period _12:10-12:20 shown in Fig. 2a, but not included in Fig. 2b. Again, this makes me question why only data from 11:10-11:45 are include in the comparison, as well as whether the fit is not as robust at high extinction.

Response: The data originally presented in Figs 2 and 4a were not from the data included in the analysis, but were presented as an example from another flight in SENEX because extinction values were nearly constant as the plane flew at a level altitude, allowing us to average over 35 minutes and get good statistics. In place of this example, we now provide as an example a profile (Fig. 3c) that was included in the data selected for the analysis. The f(RH) curve from this profile is in Fig. 7, which also includes histograms of f(RH) for all the analyzed data. Further, we provide a composite profile (Fig. 4b) produced from all of the selected profiles that shows median and variability data for extinction and f(RH) and how well the fits to the κext and gamma parameterization represent f(RH) at $\sim$70% RH. The better performance of the κext parameterization for most of the data is shown by the histograms in Fig. 7c. The histograms differ slightly from the previous manuscript because we are now including all data when the high RH channel was at 85% RH or higher (before we were filtering at 88% RH).

Comment: e) (small point: "over the time period from 11:10 and 11:45 LT" should be reworded to, e.g. "over the time period 11:10-11:45 LT")

Response: This is no longer relevant as this data example has been removed.

Comment: 6. pg 25711, lines 26-27: “This _20 % effect on f (RH) due to refractive index change for RH _ 90 % (Hegg et al., 1993) can be ignored to first order.” Here it is asserted that a 20% effect “can be ignored to first order” – yet the average 20% bias caused by using the gamma fit was earlier presented as a significant enough error to be worth exploring an entirely different fit formulation. This seems to be an inconsistency.

Response: The κext formulation does not exactly predict the change in extinction with humidity–it's only an approximation that must be used parametrically (fitted to data) rather than as an exact prediction from first principles. This issue is now discussed in Appendix section A.1., where we state, "The volume-extinction approximate proportionality in Eq. (A1) applies for an aerosol of constant refractive index, which is not the case for an atmospheric aerosol particle growing by addition of water with increasing RH (Hänel, 1976; Hegg et al., 1993). The methodology to calculate ambient extinction (Section 2.3), which incorporates the aerosol composition and size distribution measurements, can be used to estimate the effect of aerosol water on the refractive index and its impact on extinction. Using this approach, the calculated mean decrease in refractive index caused by condensed water reduces extinction by a factor of 0.81 +/- .03 for the $\sim$70% RH channel and by 0.71 +/- .03 for the $\sim$90% RH channel. Because of this effect and the rough proportionality between particle volume and extinction, Eq. A1 [the κext parameterization] is an approximation that may be used only parametrically to interpolate and extrapolate from discrete measurements on the f(RH) curve. However, it is a physically based representation of the expected functional form of f(RH), unlike alternative parameterizations."

Comment: 7. pg 25715, line 20: I think there is a typo (misplaced “r”?) toward the end of the line: “K_{chem}r”.

Response: Corrected.

Finally, we have reprocessed all of the extinction values calculated from the AMS and

size distribution measurements using the best estimate of κchem for OA of 0.05 from our measurements, rather than the 0.076 from the literature we used previously. This improves theoretical and measured comparison of f(RH) at the high humidity condition (Fig. 5).

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C12344/2016/acpd-15-C12344-2016-supplement.pdf

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