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Interactive comment on “Estimation of aerosol water and chemical composition from AERONET at Cabauw, the Netherlands” by A. J. van Beelen et al.

Anonymous Referee #1

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This paper uses radiosonde RH measurements and information in the AERONET database to estimate the concentrations of aerosol water, inorganic salts, organic carbon, and black carbon. This estimate is accomplished by:

- Assuming that all aerosols are located in a 2-km boundary layer.
- Computing a 2 km column-averaged RH from the radiosonde measurements (which allows determination of the water uptake and refractive indices of the soluble components).
- Iterate mass mixing ratios of the components, column size distribution, and RH in a model until a cost function for refractive index, AOT, SSA, volume distribution,

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and RH is minimized.

The authors assume spherical-shaped particles and use volume averaging to compute refractive indices for the mixtures of “wet” soluble components with dry components. They utilize bimodal lognormal aerosol size distributions to compute the optical properties of their modeled mixtures (rather than the AERONET distributions), and allow the fine and coarse modes to have different refractive indices (unlike AERONET). Results are “. . . compared to in situ measurements from the Intensive Measurement Campaign At the Cabauw Tower (IMPACT, May 2008, the Netherlands).”

I enjoyed reading this article and find it suitable for publication in ACP, subject to a few “major” corrections listed below. Although these corrections are important, I don’t believe that they will require tremendous effort. The “minor” corrections are less important, and left to the author’s discretion.

1 Major Issues

The authors are using Level 1.5 AERONET retrievals, seemingly with no constraints; thus, all solar zenith angles are allowed. This is a problem, because the quality of the AERONET retrievals degrades substantially at high sun (since the range of angles scanned during an AERONET almucantar is twice the SZA, retrievals at low SZA are based upon a small range of scattering angles). Hence, it is much better to “backfill” Level 2 data with level 1.5 data when low AOT retrievals are needed (since the high sun retrievals are thrown out in the Lev 2 retrievals, irrespective of AOT). Backfilling level 2 with lev 1.5 data has the added benefit of the post-field screening process, which eliminates data that reflects instrument malfunctions. The authors mention the backfilling technique on p12, but they are still showing lev 1.5 data that do not correspond to lev 2.0 size distributions. The authors need to repeat the analysis using backfilled data, in

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my opinion. They have noted that this eliminates most of the “suspect points” that they found, so their results will improve.

The OPAC values for BC in Tables 3 & 4 should be omitted, in my opinion. ? note that no group has ever measured a density of 1.0 g/cm^2 for BC, and state that the OPAC refractive index “...represents none of the possible refractive indices and should be retired.” Thus, the range of plausible densities and refractive indices for BC is not as large as the authors indicate in their Table 4, in my opinion.

Page 9:

The authors state “AERONET SSA and RI are relatively accurate for $\text{AOT} > 0.2$ but their inaccuracy is larger for smaller AOT.” I have never seen this claim. The AERONET folks make accuracy claims for Lev 2 data, which requires $\text{AOT}(440) > 0.4$, but I have not seen a validation study or sensitivity study that resulted in this conclusion. Thus, a citation or a sensitivity study is needed here.

Page 10:

Angstrom exponents great than 1 don’t necessarily indicate “... the absence of a significant coarse mode fraction.” Rather, I would say that the fine mode fraction dominates these cases, as a substantial coarse mode fraction can exist when $\text{AE} = 1$.

Page 17:

The authors point out here and elsewhere that the correlation of RRI with radiosonde RH is low, but it is not clear to me that the relationship between these parameters should necessarily be linear (is there a reference somewhere?). Sure, there should be an inverse relationship for a given aerosol hygroscopicity, but what if the relationship contains an exponential (like $f(\text{RH})$, for instance)? How would that affect the correlation? It would be interesting if the authors computed the relationship between RH and RRI, as I have never seen this (and they probably have all the pieces in their model to do this). Barring that, the authors should at least mention that they don’t know if the relationship is linear. I encourage the computation, though, because I am sure that

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many folks haven't seen it.

2 Minor Issues

The model is allowed to choose particle radii up to 50 μm , but AERONET only provides radii up to 15 μm . Does the model ever choose substantial populations of particles larger than 15 μm ? A couple of sentences on this topic would be interesting.

Also, the model is allowed to chose refractive indices for each aerosol mode, whereas AERONET reports a single refractive index for both modes. Presumably, the AERONET refractive indices are generally between the modeled values for the two modes. Do the RRI vary substantially between the modes? If so, which mode has lower RRI (which could indicate lower higher water content). Some discussion on this topic would be nice.

The first paragraph of Section 2.2.2 on page 10 ("Optimized solutions...") should be moved just above Section 2.2.1, in my opinion. That way Section 2.2 will start with a nice lead-in, and folks won't wonder why a main section entitled "IMPACT observations" begins with AERONET.

Page 3:

The authors stats "Satellite remote sensing, on the other hand, provides daily measurements of aerosol optical properties (e.g., aerosol optical thickness, refractive index) and aerosol size that cover the whole globe." Citations indicating which satellites obtain these properties would be appropriate, here.

Page 9:

The authors state: "The uncertainty in the AERONET volume distribution is 15% between 0.1 and 7 μm , increasing up to 100% at the distribution edges." A citation is also needed here. These numbers probably reflect the sensitivity study of ?, but the

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errors at the distribution edges may have been corrected when the spheroids of ? were incorporated into the retrievals.

Page 13:

"The daily cycle of the computed aerosol mass is opposite to that of the observations, with the former showing an increase and the latter a decrease during the day." – presumably, the authors mean "surface" observations.

Page 17:

The authors state that the AERONET RRI shows very large variations during the course of a single day... this could be caused by poor retrievals at low SZA. Fixing the major issue above may resolve this problem.

References:

Bond, T., and R. Bergstrom (2006), Light absorption by carbonaceous particles: An investigative review, *Aerosol Sci. Technol.*, 40(1), 27–67.

Dubovik, O., A. Smirnov, B. Holben, M. King, Y. Kaufman, T. Eck, and I. Slutsker (2000), Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) sun and sky radiance measurements, *J. Geophys. Res.*, 105(D8), 9791–9806.

Dubovik, O., et al. (2006), Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust, *J. Geophys. Res.*, 111, D11208, doi: 10.1029/2005JD006619.

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