Atmos. Chem. Phys. Discuss., 9, C7304–C7311, 2009
www.atmos-chem-phys-discuss.net/9/C7304/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comparison of in situ and columnar aerosol spectral measurements during TexAQS-GoMACCS 2006: testing parameterizations for estimating aerosol fine mode properties" *by* D. B. Atkinson et al.

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Received and published: 19 November 2009

We thank the anonymous reviewers for their general support of this paper and their considered and helpful comments. We think that the draft is significantly improved by the inclusion of their specific suggestions and consideration of their general comments. In particular, we have tried to minimize the use of non-standard acronyms, have tried to be more quantitative in the comparisons between datasets and more consistent in the general statements about derived parameters (for example, the admittedly ill-defined border between the å values expected for coarse mode-dominated vs. fine mode-dominated aerosols was moved to 1, as suggested), and finally have tried to state more clearly the underlying motivation for this work – viz, more widespread availability of multi-wavelength extinction and/or extinction-related optical measurements seems likely in the future, and the extension of those measurements to provide information about the anthropogenically-focused fine aerosol mode should provide significant dividends. We believe that this last statement is true even given the relatively large potential sources of error associated with some of the extracted values and the possible violation of some of the underlying presumptions (for example the concept of a single log-normally distributed fine mode) because of the dearth of measurements of size distributions by either in situ or remote measurements.

Anonymous Referee #1

Please note that cited line numbers refer to line numbers in the resubmitted manuscript.

"This is a good paper that utilizes a rich dataset, but I found it difficult to maintain focus while reading it. The authors compare results from using many different measurement techniques, but it is not clear why they are doing this... That is, what is the advantage of using in situ extinction measurements to ascertain the fine mode aerosol contribution, when there are other in situ measurements that do this better?"

We have added a clarifying statement on lines 72-77 of the most recent manuscript submission that points out the utility of being able extract fine mode specific physical and optical information from extinction measurements.

"Part of the problem is that there are so many non-standard acronyms. As a reader, I found myself repeatedly converting cryptic acronyms to something more meaningful. For instance, “AERONET SSRI” should simply be called AERONET that would alleviate the burden on the reader to memorize an acronym that he will probably never see again. Yes, readers are intelligent and can do these conversions, but most of us are too busy already and would rather have things spelled out."

ACPD
9, C7304–C7311, 2009

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We agree that the use of acronyms was excessive and we have cut back a little to improve the readability, including the example cited by the reviewer.

"Issues: On page 17468, line 15, the authors state: “values of Angstrom Exponents near to or less than zero pertain to large (coarse mode) particle sizes, while larger values are produced by fine mode particles.” but on page 17479-50 they state: “For period 1 (coarse mode dominated), the average value of alpha_ep for the sub 1_m data is 1.5, whereas the alpha_ep value for sub 10 _m is around 0.8.” This is a bit of an inconsistency, as 0.8 is greater than zero. On page 17468, the authors should state that Angstrom exponents less than *one* are dominated by the coarse mode, and that values greater than 1 are influenced by the fine mode as well as the coarse mode (i.e., it is not a hard cutoff)."

Agreed. One is a better value to use, but it clearly is not an absolute. We adjusted the text (lines 63-65) to reflect this remark.

"Page 17475, line 2: For coarse-mode dominated aerosols, alpha’ is <= 0. Although this is generally true, it is not always true. The authors should change this statement a little bit to reflect that alpha’ can be less than zero for some size distributions that are dominated by the fine mode."

This is a good point and we have clarified this section of the text to indicate that it also applies to very small, absorbing, fine-mode particles (lines 243 and 244).

"Page 17481, line 4: The authors state that the agreement is satisfactory (a subjective term), but don’t state why. Some quantitative comments would be nice, like: if the black line is the “truth,” then eta is 25% too low for period 1, and is often off by +/- 15% during period 2, etc."

Because eta and SMF are not really the same thing, it is difficult to make an objective (or even subjective) statement about which is the “truth”, but we would note that we have tried to be a little more quantitative about the comparison (mean values for the

ACPD

9, C7304–C7311, 2009

two periods are given for comparison) and we have added a statement to note the likely source of errors in eta – the potential sources of error in SMF (RH effects, particle bounce, etc.) are better known and documented (lines 407-414).

"Page 17481, line 7: The authors state that eta is expected to be smaller than SMF, but fig 6 shows the opposite. how come?"

What we meant to say was that there are reasons to expect eta to be smaller than SMF, which is generally true in Fig. 6, except where eta goes to unity. Examination of the DMPS data in Fig. 3 implies that this (eta = 1) may well be closer to the correct answer during this period, so the lesser values of SMF may be an artifact of the aerosol size selection, as noted in the revised text. Again the point here is that both eta and SMF have problems and neither should be regarded as the right answer.

"Page 17482, line 4: Again, quantifying "satisfactory" is important. Also, are the 24-hour averages day and night? AERONET is daytime only, so how does this affect the comparison? Additional statistics would strengthen the paper, perhaps a table with correlations, slopes, intercepts, biases, etc. of all the methods vs. the SMPS or DMPS."

Agreed, satisfactory is a nebulous word, so we removed it. The 24-hour averages are day and night, but the intensive parameters reported in this paper vary more slowly than extensive properties usually do (as shown in many of the figures in this paper) so it is appropriate to make longer term comparisons. One of the criticisms of AERONET that motivated this work is that it can only make measurements on clear days and thus does provide a sparser data set than might be desirable. The agreement between the surface extinction measurements and AERONET implies that the new generation of low cost extinction instruments could help to fill in this data record. Several of us agreed that a table of statistics would be helpful, so we have included it, although it would have been misleading to perform regressions vs. the DMPS because that would have implied that it was the preferable way to obtain Reff,f, which is not what we are

ACPD

9, C7304–C7311, 2009

Interactive Comment

demonstrating in this paper. (All of the instruments have demonstrable problems.) The table in the revised paper shows unbiased regressions between all of the different measures of Reff,f.

"Finally, some comments on the agreement/disagreement of the column vs. surface measurements would be appropriate."

There are some statements to this effect in the paper, but because we intentionally selected periods where the AOD was strongly dominated by near-surface mixing layer extinction, we are not in a good position to comment on the magnitude of the differences.

"Despite these many comments, I believe that this is a good paper worthy of publication. These comments can easily be addressed with relatively minor changes in wording."

We appreciate this constructive and supportive comment.

Anonymous Referee #2 Received and published: 18 October 2009

Please note that cited line numbers refer to line numbers in the resubmitted manuscript.

"This paper presents a solid technical aspect in spectral extinction measurement. However, the applicability and benefit of using such data to infer fine-mode fraction of extinction is unclear to me. I suggest the authors to resolve the following issues before the paper is published.

This study seeks to decouple the "fine" and "coarse" particle extinction (bep) based on spectral extinction data. The algorithms used were developed for AOD. Since AOD is essentially column-integrated bep, it is not surprising that the algorithms work equally well, if not better, for surface CRD measurements (as vertical inhomogeneity is irrelevant), providing that all measurements were quality assured (which I do believe the authors did a good job). However, one knows that R(eff,f) from pure optical methods cannot be exact, since these methods do not address particle size distribution (e.g.,

ACPD

9, C7304–C7311, 2009

Interactive Comment

number of modes and geometric standard deviation), particle morphology, and refractive index without certain presumptions/constraints. The authors need to make this clear. Although it may be found elsewhere, I suggest some descriptions of underlying assumptions, such as modal size distribution (bi-model, tri-modal?), particle shape, and refractive index, of each spectral method (FMC, GSM, etc.) to be included in the method section."

We believe we have addressed this, in response to the more specific comments of the other reviewer. For example we point out that all of the spectral methods (except AERONET, which uses sky radiance measurements in addition to AOD) have to assume that the fine mode can be represented by a definable effective radius. It is interesting to note that the concept of an effective radius was advanced to reduce the inherent complexity of the fine mode(s) to a single radiatively significant parameter, so an “exact” answer seems to be a bit of a contradiction in terms.

We would note further that the SDA/FMC method was tested over a comprehensive range of refractive indices (including absorbing aerosols), mode placement and mode standard deviation (O’Neill et al., 2005 and O’Neill et al., 2003). Tri-modal effects were investigated and it was found that the fundamental SDA equations were conserved if the fine mode curvature parameters were replaced by their mean over a bi-modal fine-mode distribution (the same comment goes for a bi-modal coarse mode distribution). This latter finding is unpublished but will likely be reported in a future paper (its derivation and relevance would go far beyond the scope of the present paper).

"For example, what conditions justify Eq. (3), (6), and (7)? How the equations would change for situations that deviate from the presumptions? Certainly, such situations might not be encountered during this study, but it is important for the readers to evaluate the extrapolation of the conclusions here to other studies."

This is largely addressed in the references cited in the comment immediately above, but is sufficiently complicated that it would make this paper much more difficult to read.

A decision was made to rely on an interested reader's ability to drill down into the references, while allowing a more casual reader to get the general idea of the various approaches from this text.

"Why R(eff,f) calculation becomes less accurate as eta decreases(p 17481)?"

This is expected since the retrieval of alpha and alpha' becomes increasingly more sensitive to the progressively larger dominance of the coarse mode extinction and its contribution to the curvature. There is effectively less and less significant fine mode "signal" as eta decreases.

"Particle absorption is less dependent on size than scattering, and therefore particles of low single scattering albedo may have smaller spectral curvature, which seems to be missed by the GSM method."

Generally absorption is small with respect to extinction, as the reviewer probably knows. In those cases where absorption is significant, it would be more appropriate to use the rotated grid presented in the Gobbi, et al. paper for a non-zero imaginary refractive index.

"In addition, it cannot be very useful if the proposed spectral approach for CRD works only for unimodal size distribution in the fine mode (as it appears to be). Especially the fine-mode fraction of bep can be achieved directly by alternating between non-size-cut and size-cut inlet of a CRD at a single wavelength (e.g., 500 nm). Please explain the benefit of applying the spectral approach to CRD."

As noted above, there are problems inherent in using aerodynamic size selection too. For example, in airborne applications, this usually requires two separate measurement channels. There is also the possibility of differential transmission in the size selection and/or creep in the cut-point from RH. So the ability to back up a size selection with the spectral method is useful. If extinction instruments are deployed that do not allow size selection (for example the long-path transmissometer that measures over $\sim$1 km open

ACPD

9, C7304–C7311, 2009

Interactive Comment

atmospheric paths), the spectral methods will still be applicable. Also, the aerodynamic size selection allows the extraction of eta, but not Reff,f so the methods used here offer additional value to using a size-selective inlet.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 17465, 2009.

ACPD

9, C7304–C7311, 2009

Interactive Comment