December 5, 2015

Dear Dr. Stier;

Please find point-by-point responses to the reviewer for manuscript acp-2015-234. Changes to the manuscript include:

- Page 8, lines 20-23: Sentence modified, with the new focus on "phase function" instead of "radiances."
- Page 9, line 5: Changed "geometrical" to "absorption."
- Page 11: Removed the following sentence at the request of the reviewer:
  "(For instance, one could compute the column aerosol scattering and extinction optical depths using nephelometer and cavity ringdown measurements from aircraft profiles, and then ratio these two parameters to obtain the column single scatter albedo.)"
- Page 14, lines 23-27: Added a sentence, emphasizing the accuracy of the Maxwell Garnett effective medium approximations for internally-mixed collapsed soot clusters.
- Page 28: Added a citation (Taylor, 1982) to justify propagation of errors in quadrature. We searched many statistics books, and found that normal-distributed errors is not a requirement for RMS error propagation (as stated by Reviewer 4). The errors are required to be independent, however, which we now note.
- Removed the confusing attempt to quantify the effect of internal- vs external-mixing from the uncertainty analysis (old page 28).
- Page 29: Added a new Section 5.4 that discusses why there is always some unquantifiable uncertainty associated with any retrieval.

Most of these changes are discussed with more detail in the response to the reviewer.

Thank-you,

Greg Schuster

The authors clearly improved their manuscript following the suggestions of the reviewers. It can be published in ACP with a few minor changes.

About new Section 2:

*** On page 8 of the revised manuscript you write "... and a viable model of the radiances at angles where they are not available". In my view it does not make sense to calculate radiances at scattering angles that can not be observed. How would that work?**

We need radiances at more angles than we measure in order to compute fluxes. If the computed fluxes are consistent with independent flux measurements, then we have closed the radiation loop at the surface. We cited two papers that checked this (Schuster 2004 and Garcia 2008), but this should be done more frequently with AERONET data (in our opinion). Flux computations are also important for computing the direct radiative effect of aerosols.

At any rate, we changed that sentence to focus on the phase function at large scattering angles, rather than radiances (page 8, lines 20-23):

> Thus, the inferred size distributions and refractive indices provide the correct phase functions and radiances where measurements are available (i.e., $\theta \leq 2 \times \theta_{\circ}$), and viable phase functions at angles where they are not available ($\theta > 2 \times \theta_{\circ}$).

*** Page 9 "increases the geometrical cross section": You probably mean "absorption cross section".**

Actually, we mean both!... however, "absorption" is probably more clear to the reader, so we changed it. Thank-you.

*** Page 10 "(Simplified shapes may also be an issue for some in situ measurements that require Mie theory for calibration; e.g., optical particle sizers, nephelometer truncation corrections, etc.)": I suggest removing the brackets and moving it to where you discuss the validation efforts.**

We would like leave this one here and include a sentence on this topic in the new Section 5.4 as well. AERONET has been receiving increasing criticism at scientific conferences because of the shape assumptions. The point of this paragraph is that we recognize that simplified shapes in the retrieval model can cause errors, but that it is important for readers to know that similar assumptions are required for some in situ measurements as well.

*** Page 11 I would remove "(For instance, one could compute the column aerosol scattering and extinction optical depths using nephelometer and cavity ringdown measurements from aircraft profiles, and then ratio these two parameters to obtain the column single scatter albedo.)" or merge it with the previous sentence.**

OK...thanks. We have removed the parenthetical phrase.

About new paragraphs in Section 5.2:
*** How is mass absorption cross section defined here? Absorption per soot mass?**

We removed that paragraph because it was difficult to follow, and it didn't really quantify the effect of external mixing on the retrieval anyways. We provide discussion on this topic in the new Section 5.4.

*** Using RMS for calculating the uncertainty assumes that the underlaying uncertainties describe normal-distributed and independent quantities. This is surely not fulfilled here, so that the calculated RMS uncertainty of 62% might still be an underestimation of the true uncertainty. However, as a precise uncertainty analysis is hardly possible with the available data sets, I'm happy with the uncertainty analysis presented by the authors. It is probably not too far away from the true uncertainties of the approach.**

Thank-you for this comment. We agree with your assessment that RMS error propagation is not really adequate for retrieval algorithms, but that there are not any other viable approaches. Some of the wording in the previous draft was difficult to follow, too, and probably not really appropriate anyways. Hence, we've attempted to articulate some of the issues associated with applying error analysis to retrieval algorithms in a new Section 5.4.