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Discussions

Interactive comment on "Evaluating Model Parameterizations of Submicron Aerosol Scattering and Absorption with In Situ Data from ARCTAS 2008" *by* M. J. Alvarado et al.

Anonymous Referee #1

Received and published: 27 February 2016

This paper describes a closure study that examines the performance of aerosol optical property treatments represented by several models when driven by observed aircraft measurements. Given that climate models have uncertainties in simulating aerosol radiative forcing and chemical transport models are sensitive to radiation via photolysis rates, this study is important in highlighting deficiencies in models that could be remedied in part by adopting more state-of-the-science treatments of aerosols. The methodology employed is similar to previous closure studies, but as far as I know the models in question have not been confronted with the type of measurements before. Despite the importance of the results of such a closure study, I have several concerns regarding specific details of the approach that affect their conclusions.

Major Comments:

1) A number of times in the manuscript the authors note that computational expense of optical properties as being a reason for simpler approaches employed. It would be useful to be more specific and actually quantify the differences in computational expense of the treatments in the four simpler models versus the ASP model. I have used Mie calculations in 3-D models for many years, and I have never thought of it being a large computational burden compared to other components of a chemistry model. My impression that one reason for many models not adopting new treatments for optical properties is that the scientific community has focused more effort on "hot topics" (e.g. SOA) that overshadow old topics that are viewed as closed and get relatively little attention. In addition there are "simplified" Mie calculations described in the literature.

2) In section there is about a paragraph of information to describe the optical properties used by the four global models used in this study, but I did not get enough information to understand how they compute aerosol optical properties as well as understanding the differences between models. The material seems out of balance with the material presented for ASP. There are two pages of information describing the entire ASP model that has little to do with aerosol optical properties and is really not needed later in the manuscript. I assume that much of the background material on ASP has been described elsewhere and could be greatly shortened. I suggest the entire section be revised to better communicate the methodology of the optical properties for all five models.

3) There is a lot of scatter associated with closure study for absorption that may have nothing to do with problems in the optical property treatments used in the models. I strongly suggest the authors compare the time series of absorption and black carbon and determine how well those two quantities correlate, and include some subset of this analysis in the paper. One possible issue is that the PSAP may not work well for low concentrations of BC, and if that is the case those data points should not be used in the closure analyses.

4) It would have been useful to propagate some of the measurement and optical property treatment uncertainties into the closure study. Considering the consequences of the measurement uncertainties, and some are no small, are particularly important since the point of the study is to assess how well the model treatments perform.

Specific comments:

Page 2, lines 21-22: "Adding in situ size distribution …" sounds awkward to me and should be rephrased. I think what the authors are trying to say is that ASP accounts for aerosol size distribution, but the other models treat aerosols as bulk species with no size distribution? Or is the difference fixed versus variable size distributions?

Page 2, lines 24-25: "mixing state" is used here when I think the authors mean mixing rule (as said earlier on line 16). Throughout the text the authors then seem to suggest that these mixing rules are mixing states – and the authors should use language that does not confuse readers. To me whether a model treats complex mixing states has to do more with just the optical properties – it is a fundamental part of how aerosol thermodynamics and dynamics are handled. Simple aerosol models that treat bulk aerosol (either in the simplest external or internal states) can manipulate model parameters using mixing rules (e.g. changing from external state to shell-core) even when that model is not simulating aerosol properties that way. In addition, there are only a limited number of mixing rules that do not reflect the continuum of mixing states.

Page 3, line 30: The authors state that the direct effect is uncertain which is true, but the uncertainties in the indirect effect are far larger and are of more concern the climate modelers.

Page 4, line 9: Change "intensity" to "expense".

Page 4, lines 29-31: Those conclusions are drawn using data collected during dry conditions and my not apply under higher RH.

Page 5, lines 14-16: I assume that a combination of these factors also contribute to the

errors?

Page 11, line 18: The authors mention the UHSAS instrument, which is an optical counter measurement that depend on an assumed refractive index (non-absorbing) that may not be representative of the ambient conditions being measured (see Kassianov et al. Atmosphere, 2015). But the authors do not describe whether the ARCTAS data has been corrected for this artifact. The authors should determine if the data has been corrected and describe some of the uncertainties in the measurements that will affect their closure study.

Page 12, lines 1-2: A reference is needed for these measurement uncertainties. Measurement uncertainties quoted in the literature for the AMS instrument varies widely, unfortunately.

Page 12, line 28: The statement regarding the aerosol mass to be dominated by OA and BC is strange. BC is usually very small, except in the center of biomass plumes. I would still expect sulfate to be a large fraction of mass. In fact, the AMS measurements should tell you whether there is significant sulfate (or nitrate and ammonium too) or not.

Page 13: end of Section 3.2: I would like to see a figure showing the merged size distribution and the individual size distributions from the SMPS, UHSAS, and APS. The critical part is merging these distributions where they overlap. I suspect that at individual times, the overlap might have large disagreements – but that when the data is averaged over a period of time those disagreements likely become smaller.

Page 14: Section 3.3: More discussion is needed regarding the uncertainties in the scattering and absorption measurements. For PSAP, there have been a number of studies pointing out some of the flaws with the instrument.

Page 17, lines 1-8: More description is needed on how the aerosol composition is used. I understand the size distributed AMS data is either not available or it may not be desirable to average the optical property measurements over an hour period. But

the size distribution for organic matter, sulfate, nitrate, and ammonium may not be the same. If they are assumed to be the same this is an important piece in the methodology to mention.

Page 18: lines 23-30: I do not see where data for the 450 nm channel are presented.

Page 20, lines 3-16: There are studies that suggest that uncertainties in SSA needs to be very small to have confidence in radiative forcing estimates. The results from this study show that even with the observations we cannot achieve this (large amounts of scatter). So what does this mean for climate model calculations that will have even larger errors in size, composition, and mass? Some perspective on SSA accuracy needed for climate models based on from other studies needs to be discussed.

Page 22, line 7: The authors suggest that BC is expected to be externally mixed for fresh smoke. I suggest the authors actually provide evidence for this. A number of studies have shown that SP2 can be analyzed to produce coating thickness as a function of size. If the authors are correct then the coating thickness should be small to zero when the aircraft is sampling in fresh smoke, and larger thickness further downwind. Is coating thickness available from the ARCTAS SP2 measurements

Page 22, line 13: I do not think enough proof is provided regarding mixing state in the observations.

Page 22, line 26: Table 7 is presenting the results by mixing rules, not mixing state.

Page 24, line 12 and 20: I do not think the model does a reasonable job in absorption, contrary to what the authors state in line 12. Then in line 20 the authors state that there should be improvements in absorption – a contradiction in this paragraph.

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