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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Remote sensing of soot carbon – Part 1: Distinguishing different absorbing aerosol species" *by* G. L. Schuster et al.

Anonymous Referee #2

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1 Overview

This paper discusses the interpretation of remote sensing data, especially AERONET, to extract concentrations and relative fractions of absorbing species. Improved separation within this widely used data set will benefit the community by providing observational data for understanding atmospheric concentrations of aerosols and for evaluating models of black carbon, brown carbon and dust. To date this separation endeavor has been done as part of modeling studies, often relegated to appendices, and not carefully evaluated. This type of work is a welcome addition to the literature. The logic behind the general approach is sound. The paper is well written and the assumptions are clearly stated. I have some questions which were not fully answered by the paper's

first version. I recommend publication if these can be addressed.

2 General comment on the field

Given the wide use of AERONET retrieved data, it is really surprising that so much effort has been put into its theoretical interpretation, and so little effort into confirmation with optical and chemical measurements. I realize that this is not easy, because AERONET senses the column, which is difficult to compare with in-situ measurements. This isn't authors' fault and not a reason to criticize the paper, but perhaps authors could make some comment on this state of affairs.

3 Major questions

3.1 Consistency with radiance field

I start with the end: where authors critique the procedures in other papers. They state: "Reformulating the model assumptions in a manner that is not consistent with the original retrieval can produce results that are inconsistent with the radiance measurements." - page 13629, line 8. This is an important point and should probably appear earlier in the paper. Any proposed size and refractive index, should be consistent with the entire radiance field. So a dust coarse mode plus BC fine mode (as assumed in Bond paper) could not reproduce the radiance from which the size distribution is derived, and there could be a similar problem with Bahadur and Chung assumptions. In contrast to representations that violate the retrieval assumption, authors propose to keep the single refractive index and adjust the components to match it separately in coarse and fine mode. ("Single" here means "across the two modes"– of course each wavelength is

different.) I discuss the problem in context of the two-mode model because it is easier, but the assumption by Bahadur or Chung would have something similar.) This assumption is now consistent with the one in the retrieval and thus the proposed aerosol would produce the radiance field observed. In case the dust cannot match the refractive index in the coarse mode, some sC or BrC is added to this mode. Regardless of whether this assumption is consistent with retrieval, the most important question is whether the single refractive index is physically reasonable. Would one expect identical refractive index in the two modes? It seems quite plausible that they would differ. Nature does not conveniently add sC to the coarse mode in order to match the refractive index of the fine polluted mode. I respect the consistency argument, but physical accuracy would be more important. Authors say this is "consistent with what is found in nature" (Page 13613. Line 10) Indeed a small amount of dust is found in the fine mode, and sC, BrC in coarse mode, but there is no reason that this cross-over must create equal refractive index in the two modes. This raises the following questions:
1. The assumption of single refractive index is imposed by the retrieval designer. Is it possible to confirm whether it is accurate or is there simply not enough information in the radiances?
2. The other works may have a problem of consistency with the radiance. How bad is this inconsistency? It should be not very hard to calculate angular scattering with the different assumptions.

In order to create consistency the authors have worked out the allocation scheme in Figure 6. It seems that any retrieval requires some treatment or assumptions.

3.2 Exploration of refractive index space

Much of this analysis relies on AERONET reported refractive index (imaginary). What is the uncertainty in that report? 0.004 seems like a rather small number.

I find Figure 3 very difficult to interpret and think the labeling needs to be improved.

This is important because it is needed to understand figure 4. I think the values that go from lower left to upper right are volume fraction iron oxide. That should be explicitly stated and perhaps even marked. I get the impression that the brown shaded area is possible without any BC and BrC, but then the grey lines are introduced. Eventually after reading several times I think I understand the meaning: a large range of refractive index is possible from dust, and more than covers the refractive index space possible by varying BC and BrC. The BC, BrC lines are included to make this point only. If so it should be stated. Whether I have understood it or not, the figure is not easy to understand. The inclusion of the grey lines suggests at first that the refractive index space includes the BC, BrC.

Actually I suggest it would be better if all figures were labeled according to their message, e.g. Fig 3 "Refractive index space for dust (brown) and BC-BrC mixtures (gray lines)", Fig 4 "Refractive indices observed for pure dust and biomass burning"

Now if my interpretation of Fig 3 is correct, it is difficult to see the possibility of any retrieval at all. A large variation of BC, BrC volume fraction lies within a small variation of dust volume fraction.

Fig 4 – pure dust observations are used to identify a dividing line between dust and biomass burning. Despite my questions here, I find authors' choice of this division (k=0.0042) supportable.

Authors later assume relative fractions of hematite and goethite (0.4%, 0.6%) but don't discuss it. The implication from the observations should be discussed. Wouldn't this vary at other locations?

Section 2.3: Authors demonstrate cases when biomass burning and dust are mixed. These are the most difficult cases and authors propose an algorithm (Figure 6). A major assumption is the rightmost branch in the figure. The volume fraction of iron is set to 1

3.3 Fixed dust assumption

One of the challenges is separating aerosol types when they are mixed. For a certain level of absorption the authors fix the volume fraction of iron. Possibly, there is not enough information in the retrieved data to do better than this. But this assumption needs to be considered in the interpretation. Authors state “the coarse mode iron oxide mixing ratios are still steady throughout the year (at 1.1–1.3 %).” (page 13621: line 12) But they have also said that the fixed-dust branch of interpretation had to be used for most of the biomass-burning sites in South Africa. Wouldn’t one expect to find a constant iron oxide mixing ratio if that branch was often used?

I would rather see a discussion of what iron oxide mixing ratios vs sC/BrC are consistent with the observations. If this answer is a large range, that is also good to know because it implies that one cannot extract the separation from the data as given.

4 Other comments

Page 13621 – Discussion of BrC/sC ratio. The distinction between BrC and OC should be brought in the beginning of the paper. It seems not-absorbing OC is part of the host.

Estimates of BrC/sC ratio, and comparison with expectation. Authors cite Chen and Bond (2010) and not any other material for this expected ratio. I suggest review papers such as Andreae and Merlet (2001), Reid et al (2005). In fact I did not find any measurements of sC in Chen and Bond paper.

I appreciate the analysis of uncertainties. Fig 11 and 12, since we don’t know which of these is right, I suggest we don’t call it “Relative bias” but perhaps “difference.” or “Ratio with baseline retrieval.” One should also be careful about representativeness of the refractive indices used. Some are for biomass burning and some are for water-

soluble which is just a part of the aerosol. The latter would not be expected to represent atmospheric aerosol.

Section 5 – Other than my previous comment, I do not clearly see the message of this section. I think authors mean that all quantities are retrieved using the refractive index, which is part of the retrieval, so they all require the same assumptions. Both AAOD and sC loading need the refractive index. The question not addressed here (or elsewhere) is the robustness of these different quantities. In previous sections, authors show a variation in sC volume fraction with differing refractive index but did not calculate sC AAOD. Can some comment be provided on this?

5 Minor comments

Page 13610 Lines 1-3. Reasoning seems convoluted. Isn’t sC reduction proposed because of its warming, not because it reduces aerosol cooling?

Line 22 “link between emissions, concentrations, AAOD is not straightforward” – I think the link between concentrations and AAOD is not too bad, but maybe the measurements also play a role in the comparison?

Page 13614 Line 1 “two absorbing inclusions” – Does this refer to one inclusion in the fine mode and one in the coarse mode?

Figure 10 – The caption should be descriptive of the content. “Colormap” is not descriptive.

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