*Reviewer comments in **bold**.*

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.

Thank-you for the thoughtful review. We have made extensive changes to the document, mostly in red text. Long sections of red text are difficult to read, though, so we just indicated that they are "new" where necessary. The typeset ACPD .tex document does not include line numbers, so we refer to line numbers in the original draft where this is applicable.

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.

The new Section 2 discusses the difficulty of measuring aerosol properties at ambient relative humidity, and in conditions suitable for Level 2 processing. It also provides some new citations.

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?

Indeed, the AERONET Cimel does not provide enough information for automated retrievals of two or more external mixtures. We've added the following paragraph to Section 2:

The AERONET algorithm also assumes that all particles in the atmosphere have the same complex refractive index (regardless of size), which is equivalent to assuming that all particles have identical composition (and all aerosol species are internally mixed). This assumption is necessary to achieve a unique solution, and forces the absorption to be spread over all retrieved particle sizes, even if the absorption really occurs in only the smallest particles. The repercussions of this assumption are discussed in Schuster et al. (2015).

The citation is Part 2 of this paper, which is now online.

We also recognize that AERONET redistributes fine mode absorption to the coarse mode on page 13619, line 22 of the original draft:

Although carbonaceous aerosols are sometimes observed attached to coarse mode dust (Derimian et al., 2008; Hand et al., 2010), it is also likely that the AERONET product redistributes some fine mode absorption to the coarse mode (Schuster et al., 2015).

The phrase "consistent with what is found in nature" is a parenthetical element, meant to convey that it is not ridiculous to include dust in the fine mode or carbonaceous aerosols in the coarse mode. We've changed the phrase to "(which is qualitatively consistent with what is found in nature)."

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.

Well, the AAE approaches (Chung, Bahadur) can not be linked to aerosol microphysics, so these computations can not be performed. That is, one can not deduce particle size distributions and refractive indices that correspond to the component AAODs in those papers; hence we can not compute the corresponding radiances.

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The Bond (2013) approach does provide a microphysical link between dust and dust AAOD. However, the microphysical link to sC is unclear. That is, Bond (2013) fills up the coarse mode with dust. Does that mean that all of the sC is in the fine mode? If so, the refractive index provided by AERONET would have to be adjusted to account for this extra absorption. Since Bond (2013) does not provide these refractive indices, we would have to effectively “retrieve” the Bond (2013) fine mode refractive indices by iterating until we matched their AAODs. That requires a significant coding effort that is beyond the scope of this paper. However, we feel that it is important to emphasize that AERONET’s strength lies in the link to the radiation field, and that authors should be cognizant of this when they analyze AERONET retrievals.

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.

Dubovik et al 2000 assessed imaginary refractive index uncertainty at +/- 50%; we have added this to the uncertainty analysis. Yes, 0.0042 is small, but imaginary refractive index is never large in the Level 2 AERONET product (none of the whiskers in Figure 5 extend beyond 0.04). The climatological value for dust is 0.0012-0.0015.

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.

Yes, this is stated on page 13615, lines 24-26 of the original manuscript; it is also stated in the caption. We slightly modified the caption in the revised text -- hopefully this helps.

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.

We are sorry that you had so much trouble with this section. Yes, the refractive index space of dust and carbonaceous aerosols overlap. The point of figure 3 is that we can uniquely retrieve the absorbing minerals (hematite and goethite) or the carbonaceous aerosol fractions if carbonaceous and dust aerosols are not mixed together. When the two components are mixed, we need additional constraints. We use figure 4 to define these constraints. We have combined the old sections 2.1.1 and 2.1.2 and relabeled the combined section (now 3.1.1) as “Imaginary refractive index space for dust and carbonaceous aerosols.” We have also added some new introductory text to the subsequent section (new section 3.2). This will hopefully alert the reader up front that we will be discussing several aerosol absorbers.

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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"

We have changed the first line of the caption to "Theoretical imaginary refractive index space occupied by dust and carbonaceous aerosols." Titles indicating "pure" dust and biomass burning are included above each panel in Figure 4.

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.

Yes, there is a large overlap for the theoretical computations of imaginary refractive index. However, we also have fine/coarse mode separation, the k_rnir < 0.0042 constraint for dust, and the BrC/sC < 15.2 constraint. This is discussed on page 13612+, lines 24+ in the original draft, and in subsequent sections.

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?

Very good point. We've added the following text to the end of Section 3.3:

Our choice of 1% free iron in Branch D of Figure 6 is based upon climatology for pure dust at the West African and Middle East sites. We retrieve median hematite and goethite fractions of 0.39% and 0.62% for pure dust in West Africa, 0.34% and 0.58% for pure dust in the Middle East, 0.38% and 0.62% when both these regions are combined. This corresponds to $k_{440/674/870/1020} = 0.0036/0.0014/0.0013/0.0013$ *for pure dust in the combined dataset, which is also consistent with Kim et al. (2011).*

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 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?

Good point. We've added the following discussion to Section 4, and modified Figure 9 accordingly:

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The lower left panel of Fig. 9 indicates that the volume fraction of free iron remains relatively constant in West Africa throughout the year (1.4–1.7 %, as shown by the solid maroon line). However, the signal is somewhat stabilized because we use climatological mixing ratios for iron whenever the retrieval is contaminated by carbonaceous aerosols, which constitutes 24% of the retrievals in West Africa (i.e., "branch D" of Figure 6). Hence, we also show the monthly averages that are obtained by omitting the contaminated retrievals (dashed maroon line in the lower left panel). Note that both monthly averages produce nearly identical free iron mixing ratios in the absence of biomass burning aerosols during the summer months. Monthly-averaged free iron mixing ratios obtained during the winter biomass burning season increases when we omit retrievals that require branch D of Figure 6, though, peaking at 2.3% in December. This is because omitting retrievals that require branch D results in a heavier weighting of the retrievals with k_{rnir} greater than the median value of pure dust (i.e., $k_{rnir} > 0.0013$ and $k_{rnir} < 0.0042$). Thus, omitting the branch D retrievals causes the monthly iron volume fractions to increase during the biomass burning season, and more closely mimic the monthly carbonaceous aerosol signals. Hence, it is likely that some of the remaining retrievals are still contaminated with carbonaceous aerosols during the biomass burning season. Finally, we note that branch D is called for 98% of the retrievals in southern Africa, so the solid maroon line in the lower right panel indicates very little seasonality because it is dominated by climatology.

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.

Indeed, we used the literature values for the component mixing ratios to guide our choices of component refractive indices. Iron oxide mixing ratios found in the literature are discussed on page 13624, lines 27+ of the original document. BrC/sC ratios are discussed on page 13621, lines 19+ of the original document.

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.

We did this on line 9 of page 13609 in the original draft.

It seems not-absorbing OC is part of the host.
Yes. Everything that is not BrC, sC, goethite, or hematite 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.

Thank-you for the citations. We used the Chen and Bond citation because the OC/sC ratio of 3-12 that they quote is commonly observed elsewhere in the literature. We've added the Reid citation, but did not find any OC/sC ratios tabulated in Andreae and Merlet.

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

*Reviewer comments in **bold**.*

"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.

Very good points. We've changed Figs 11 and 12. We included WSOC in those figures because Chen and Bond (2010) concluded that "Water-soluble OC absorbs some light," and a significant portion of organic aerosols in the atmosphere are water soluble. However, we agree that WSOC is not appropriate for representing BrC.

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.

Yes, this is what we are trying to convey. This section was unclear to all of the reviewers, so we have omitted it from the revised version. Portions of Section 5 are now incorporated into Section 2 and elsewhere.

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?

We have added substantial new text to the error analysis (now Section 5), including the uncertainty in the AERONET retrieval as well as uncertainty associated with externally mixed sC.

Not sure that we understand the question about sC AAOD, though -- how would linking sC volume fraction to AAOD make the retrieval more robust? Although sC AAOD is an important parameter, it is not equivalent to the radiative effect (and it is certainly not easier to measure than sC mass). Modelers still need the absorption forcing efficiency (AFE) in order to relate AAOD to the direct effect, and the AFE varies by an order of magnitude in the models (Schulz, Atmos Chem Phys, 2006, Table 4). Even if we throw out the two extreme modeled AFE in Schulz 2006, there is still a factor of 2 difference in AFE amongst the models.

Additionally, internal mixing of absorbing aerosols with other aerosols alters the AAOD, so one can not calculate component AAODs for mixtures that contain more than one absorbing species. (One could calculate AAOD_effective = AAOD(with) - AAOD(without) for each aerosol component in an internal mixture, but the sum of AAOD_effective for all of the components will not add up to the total AAOD.) The AERONET retrieval assumes that all aerosols are internally mixed, so we are stuck with this assumption if we want to maintain the link to the radiance field.

We don't want to get into a discussion about which parameter(s) are the most important for improving the community's understanding of sC radiative forcing, but we feel that sC mass is just as valid of a parameter as AAOD for constraining and improving global models. Ideally, it would be preferable to have both parameters so that we could check the modeled mass absorption cross-sections (MACs). We would also like to see more modelers track sC volume fractions of the total aerosols, as this parameter is likely more closely related to SSA than sC mass.

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?

Our rationale for the wording that we chose is that sC is co-emitted with other aerosols. Since sC is an aerosol, it is transported and removed via similar mechanisms as other aerosols. Thus, it has a regional distribution that is similar to aerosols, and only warms the atmosphere in those regions. There are co-emitters that cool the aerosols in the exact same regions, so overall the radiative effect of sC plus co-emitters is generally cooling (except, perhaps, when the system of aerosols is located over clouds or snow). Since sC only warms regions where aerosols are cooling the atmosphere, the effect of sC is to reduce aerosol cooling. Aren't "less cooling" and "warming" somewhat equivalent, though?

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?

Linking concentration to AAOD requires knowledge of the sC mixing state, which is usually unknown. For internal mixtures, the relative proportion of sC in the particles is also required; that is, the particle size and coating thickness must be known. Also, one needs to know the proportion of internal and external mixtures to compute AAOD from concentrations. At the present time, both the coating thickness and the proportion of coated particles are rarely measured. This state of affairs will improve as more advanced analyses of SP2 data become available.

Additionally, the community still can't compute MAC = 7.5 m2/g for external sC mixtures with reasonable particle radii and refractive indices, as noted by Bond and Bergstrom (2006). This might be caused by the complexity and variety of shapes associated with the sC aggregates, or something else.

Since we can not compute the AAOD from what we think we know about sC size, shape, and refractive index, the link is not straightforward, and we can not adequately model AAOD. Alternatively, a modeler can postulate that MAC is 7.5 m2/g for externally mixed sC, and increase the value by 50% for internally mixed sC, but that is not a terribly robust link, and it only works at one wavelength (550 nm). A different MAC is needed for the other shortwave wavelengths, and the inverse wavelength "law" for absorption only works for externally mixed particles (maybe... it probably doesn't work for collapsed aggregates).

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

Two inclusions in each mode. The words "for each mode" have been added after Eq 1. Hopefully this clarifies.

*Reviewer comments in **bold**.*

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

We rephrased the caption, which no longer contains the word colormap.