*Reviewer comments in **bold**.*

This manuscript uses AERONET direct sun and sky scan retrievals to infer differences in absorbing aerosol species. The emphasis is on distinguishing soot, brown carbon, and hematite. Figure 2 shows that there are distinguishing features. However, it is difficult to assess how quantitative the separation between soot, brown carbon, and hematite is. It could be right, but there is just not enough error analysis presented, and not clearly enough, to convince a reviewer who is familiar with the retrievals but not a full-time radiative transfer modeler. I have five comments:

Thank-you for the many very useful comments. We have now included the AERONET refractive index uncertainty and mixing state uncertainty with the other uncertainties in the revised analysis. The error analysis for size and complex refractive index was provided in Dubovik et al 2000. The error analysis for component refractive index uncertainty is provided in Figures 11 and 12. Hopefully you will find this satisfactory.

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.

1) Coated aerosols: The retrievals assume internally mixed aerosol except for the fine – coarse mode distinction. But their method of sensing the difference between soot and hematite relies heavily on the difference between red and blue optical properties. This difference is quite sensitive to the degree of internal mixing of soot. One can show with a coated sphere model, for example, that the Angstrom exponent of absorption of soot is sensitive to the coating thickness even if there is no brown carbon, and that the Angstrom exponent of BC is sensitive to coating thickness (Lack and Cappa, 2010).

Thank-you for your comment. Presumably you are talking about Branch A of the fine mode in Figure 6, regarding the soot/hematite distinction. This branch of the code is only used in 12-14% of the retrievals at the dust sites, and is never called at the biomass burning sites (p13, lines 7-10 of the original manuscript). Thus, we don't rely heavily on red/blue optical properties to separate hematite from sC. This separation is done mainly through particle size (i.e., we mainly locate carbonaceous aerosols in the fine mode, and free iron in the coarse mode). However, your argument could also apply to the sC/BrC separation in Branch B of the code (see Fig 6), which is called ~87% of the time at the dust sites, and 100% of the time at the biomass burning sites.

The retrieval of BrC does rely heavily on the red/blue differences in refractive index, but the retrieval of sC does not. The sC mixing ratio is determined by the 670-1020 nm wavelengths, where BrC does not absorb at all. Since the sC mixing ratio is determined only by the red and nir wavelengths, the red/blue optical properties do not affect the retrieval of this component. The ambiguity of available BrC refractive indices and their spectral dependence does have a strong impact the BrC retrieval; this is recognized in the top panel of Figure 12.

We recognize the ambiguity of AAE with respect to component mixing ratios, and address this issue extensively in Part 2 of this paper (which was not available until recently). The reason that AAE changes wrt coating thickness is because MAC changes wrt coating thickness, and it changes differently at the different UV-VIS-NIR wavelengths. In Figures 2 and 3 of Part 2 of this paper, we show contour plots of AAE for mixtures with a spectrally flat absorber (like sC) in a non-absorbing host (using the Maxwell-Garnett effective medium approximation and Mie theory). We note that AAE increases as the sC mixing ratio decreases (for mixed particle sizes < ~0.1 um), which is consistent with Lack and Cappa (2010), Gyawali (2009), and Bond (2001).

We emphasize that we don't use AAE for our retrievals, though -- we use refractive index. Although the AAE changes when sC is mixed with a non-absorbing host, the spectral dependence of the refractive index of the mixture remains spectrally flat. This is also discussed in our companion ACPD paper.

Please keep in mind that sC mixing ratio is somewhat related to coating thickness (except one applies to homogenous particles and the other applies to heterogeneous particles; one applies to the sC fraction, the other applies to the host fraction). Thus, we are accounting for variable coating thicknesses and variable mass absorption cross sections (MACs) with variable sC mixing ratios.

2) Nitrogen dioxide. For a manuscript that relies heavily on blue/red differences, I am left wondering what the impact of NO2 would be, since it has an absorption spectrum that can mimic brown carbon or hematite. Absorption by NO2 in the blue can be significant compared to aerosols.

As explained above, we are not relying on blue/red differences for the sC retrieval. NO2 could affect BrC and fine mode hematite, though. AERONET uses climatological values to correct for NO2 in their products, so NO2 concentrations that are significantly different than the climatological values could affect their products. This would most likely occur in urban regions, where NO2 concentrations are high and BrC concentrations are expected to be small. Thus, we've added the following sentence to Section 5.3:

Biases in the BrC retrieval could also occur whenever NO_2 optical depths differ significantly from climatological values, but we do not attempt to quantify that here.

3) Error propagation, especially systematic errors in direct and scattered measurements. I've gone back and reread the 1998 Dubovik paper on AERONET retrievals of smoke and I still do not understand how systematic errors affect the retrievals of absorption. It seems to me that the only way to tell if absorbing aerosol is present is if there is light taken out of the direct beam that does not reappear elsewhere in the sky. Absorption cannot be determined from just a phase function because pretty much any phase function can always be fit with a multimodal size distribution. If the absorption depends on comparing the direct and scattered measurements, then their relative calibration is important. But for AERONET they are calibrated separately. In addition, common-mode scale errors could lead to incorrect albedos. A photometer sensitivity change that reduced the amplitude of both the direct and sky signals would look like absorption – the sun would be darker and the light would not reappear in the sky. Maybe there is some internal air-mass consistency check that is not explained?

It is great that you dug into the old literature. Presumably you are talking about:

*Reviewer comments in **bold**.*

Dubovik, O., B. N. Holben, Y. J. Kaufman, M. Yamasoe, A. Smimov, D. Tanre, and I. Slutsker (1998), Single-scattering albedo of smoke retrieved from the sky radiance and solar transmittance measured from ground, J. Geophys. Res., 103(D24), 31,903–31,923.

That paper is different than the present-day AERONET retrievals, though. See also

Dubovik, O., and M. King (2000), A flexible inversion algorithm for retrieval of aerosol optical properties from sun and sky radiance measurements, J. Geophys. Res., 105(D16), 20,673–20,696.

Dubovik, O., A. Smirnov, B. Holben, M. King, Y. Kaufman, T. Eck, and I. Slutsker (2000), Accuracy assessments of aerosol optical properties retrieved from Aerosol Robotic Network (AERONET) sun and sky radiance measurements, J. Geophys. Res., 105(D8), 9791–9806.

We have added a rather long section that describes the AERONET product (see Section 2 of the revised manuscript). We have also added the AERONET refractive index uncertainty to our sensitivity study, and added an additional component to the uncertainty in case of 100% external mixing (see new text in Section 5.2). Hopefully that helps.

Yes, the older AERONET Cimels have separate detectors for the sun and sky measurements, and the sensors are calibrated separately. However, there is a consistency check during all almucantar scans at the 6 degree azimuth angle, where both detectors provide measurements. If the consistency check fails, the retrieval is thrown out. This is discussed on page 7 (middle paragraph) of the new draft.

Per your example, the instrument could malfunction and cause both the sun and the sky to “go dark” in such a way that the consistency check passes; in this case, the Level 1.5 processing would probably provide a poor retrieval. However, the Level 2.0 processing includes post-field calibrations that detect and account for significant changes in instrument sensitivity. The post-field processing is described in

Holben, B., et al. (1998), AERONET – A federated instrument network and data archive for aerosol characterization, Remote Sens. Environ., 66, 1–16.

Additionally, Langley sequences are attempted daily; this provides the opportunity to pinpoint discontinuities in the TOA I_0 values when post processing.

4) Similarly, there is no discussion about how assumptions in the size distributions propagate into the results. I know some of this has been discussed in previous publications, but this analysis represents a new level of detail for which new effects might become important. According to Dubovik et al., (JGR, 1998), “: : : good agreement between the measured and the fitted radiances probably results in reliable estimates of single-scattering albedo and phase function even if the microphysical aerosol model is not perfect”. Is the black carbon volume fraction similarly insensitive to the microphysical model?

The black carbon volume fractions are constrained by the AERONET imaginary indices at the red and near-infrared wavelengths. Thus, the AERONET-fitted radiances can be reproduced with the volume fractions that we deduce and the AERONET microphysical models. If all sC is internally mixed (as in the AERONET model), then the sC volume fractions are probably reliable. We address the AERONET refractive index uncertainty and external mixing in the last few paragraphs of Section 5.2, which we reproduce here:

Other biases are more difficult to assess. For instance, we stated earlier that the AERONET retrieval model assumes internal homogenous mixing for all aerosol particles; this could produce a bias in our sC retrievals if all sC particles are externally mixed, which we discuss here. Consider two systems of particles with a given amount of sC — the sC is 100% externally mixed in one system of particles, and 100% internally mixed in the other system. Since externally-mixed sC has lower mass absorption cross sections (MAC) than internally-mixed sC, AERONET will infer a lower imaginary refractive index (and AAOD) for the external mixture than for the internal mixture. The resulting lower imaginary index produces lower sC mixing ratios in our retrievals, which also increases the MAC. Thus, the sC mixing ratio will have a low bias and MAC will have a high bias when all sC is externally mixed. Since externally-mixed MAC is approximately 2/3 of internally-mixed MAC (Bond and Bergstrom, 2006), we estimate that our sC retrievals will have a low bias of 33% when all sC is externally mixed.

We are now in a position to estimate the uncertainty associated with retrieving sC mass from the AERONET products. Recall that the estimated uncertainty for the imaginary refractive index in the Level 2 AERONET products is 50% (Dubovik et al., 2000). The measured sC densities are well known (1.8 g cm $^{-3}$ *± 6 %, per Bond and Bergstrom, 2006). Thus, the RMS error in retrieving sC mass that is associated with the AERONET refractive indices, sC density, mixing state, and the component refractive indices of sC, hematite, and BrC is* $(50^2 +6^2 +33^2 +14.2^2)$*^.5 =62%.*

Of course, other errors are possible, some of which are not quantifiable. For instance, the scientific community does not have a method for modeling collections of particles with irregular and rough shapes, so we can not compute the effect of using spheres and spheroids to approximate real particle shapes.

5) I cannot comment fully on the strong statements about the Bond et al. interpretation. I do think that Figure 13 in the discussion manuscript is misleading. The uncertainty in a parameter does not depend on where it appears in the computer code. The uncertainty is the result of error propagation. Just because the AAOD is computed after (or before) the refractive index does not make it more or less certain. Indeed, the SSA, which is listed in the top line, could as easily be computed from the size distribution and refractive index and moved to the bottom line. Another way of putting this might be to consider my previous comments. Would a change in relative calibration of the direct and sky measurements affect the refractive index or the AAOD more? What about a change in assumptions about size distributions? Is the refractive index or AAOD more sensitive? What about coating thickness on soot?

We agree that the uncertainty in a parameter does not depend upon where it is located in the computer code -- that was the main point of the discussion. Apparently our presentation was poor, though, because all of the reviewers missed our point. We were not claiming that refractive index is better than SSA or AAOD -- we were pointing out that it is not worse.
At any rate, we have moved most of the statements about the Bond interpretation to Section 2 (at the request of Reviewer #2). We have improved the wording, so that we are now discussing how AERONET works in general terms, and provide additional discussion about the AERONET model assumptions. We also include the uncertainty associated with 100% external mixing in the discussion (Section 5.2). The main point that is often missed in the literature is that all

particles are always internally mixed in the AERONET retrieval, and this affects how we can interpret the data.