*Reviewer comments in **bold**.*

This paper presents an approach to separate several types of absorbing aerosols from AERONET products of size distribution and refractive index. The main advantage of the proposed method is that its outcome is consistent with the size distribution and refractive index from AERONET and thus also with the radiation fields measured by the sun photometers. The paper is well written and the method is well explained. The main issue is that the uncertainties of the complete procedure (AERONET + proposed method) are not well discussed. I favor publication of this paper in ACP considering the remarks below.

Thank-you for your thoughtful comments. We have added the AERONET refractive index uncertainty to the analysis. We have also estimated the uncertainty associated with the possibility of 100% externally mixed sC.

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 General remarks

The proposed approach separates the different components such that the size distributions and refractive indices are not changed compared to those given by AERONET. This results in consistency with the AERONET model and the radiation field. Though this approach is reasonable given the available AERONET products, questions remain about the uncertainties of the results and how well AERONET retrievals themselfs perform when complex aerosol mixtures with size-dependent refractive indices are measured.

For example, what happens if there is a strongly absorbing fine mode and a very weakly-absorbing coarse mode? One could expect that AERONET reports some "average" refractive index, with the consequence that the absorption by the fine mode is underestimated and the absorption by the coarse mode is overestimated (which however might be (over-)compensated by branch D in Fig. 6 if there is a sufficiently large fraction of fine mode absorption).

Yes, AERONET reports an intermediate refractive index when there is a strongly absorbing fine mode and a weakly absorbing coarse mode (see Dubovik et al, 2000, Fig 10). So yes, fine mode absorption is underestimated and coarse mode absorption is overestimated in these cases.

Some authors uniformly use a climatological value (k_dust) to compute AAOD for coarse mode dust of the AERONET retrievals. This results in too much dust AAOD whenever $k_{aeronet} < k_{dust}$ (i.e., the computed AAOD for the coarse mode is greater than the AERONET-retrieved AAOD for the coarse mode), and therefore too little sC AAOD. If AERONET climatology is used for k_dust, then the overcompensation will occur ~50% of the time.

However, this is not an issue with Branch D in Fig 6, because that branch is only called when $k_{rnir} > 0.0042$ in the AERONET database. Since $k_{rnir} \sim 0.0013$ for the dust that we use in Branch D (and $0.0013 < 0.0042$), we are able to add sC to all cases where Branch D is called. Thus, we are able to remain true to the AERONET refractive indices.

*Reviewer comments in **bold**.*

To answer another way, one could say that AERONET "redistributes" absorption from the fine mode to the coarse mode in the case that the reviewer describes. From an aerosol microphysical standpoint, this generally means that sC is getting redistributed from the fine mode to the coarse mode. Our algorithm acknowledges the redistribution of sC, but does not "put the sC back" into the fine mode; rather, we allow the sC to remain in the coarse mode. This likely means that the distribution of sC between the fine and coarse modes in our retrieval is incorrect, but that the total sC that we retrieve has a link to the radiance field.

The reader is left alone with the task to estimate the uncertainties of the final results. The sensitivity study in Section 4 is good, providing an estimate of the uncertainties due to the assumptions on the component's refractive index. Uncertainties due to the decision tree (Fig. 6) are partly covered by the statistical analysis, but should be discussed more quantitatively in the final paper. The following uncertainties are not discussed or quantified:
(a) Uncertainties due to uncertainties of the AERONET-derived refractive index
(b) Uncertainties due to limitations of the model (here the size-independent refractive index is most relevant)
To cover a), I suggest that the authors add a sensitivity study on the effect of the uncertain AERONET refractive index on the type separation (by varying the AERONETderived
refractive index within the expected uncertainty) to give an estimate of this

All very good points...

a) Dubovik et al (2000) estimates a 50% imaginary index uncertainty, and Bond and Bergstrom (2009) estimate a 5.5 % uncertainty in sC density. We have now included the AERONET refractive index and the sC density in the error propagation. We prefer this path, because the component densities and component refractive indices are independent parameters, and they are independent of the AERONET retrievals. Thus, we use RMS error propagation.
b) In the revised paper, we now refer the reader to the accuracy assessment presented in Dubovik et al (2000), which covers this exact issue (see their Figure 10). This paper indicates that the intermediate refractive indices inferred by the retrieval model produces the same single-scatter albedo as the externally-mixed forward model (at least in those test cases). That paper also covers other limitations of the AERONET retrieval model. We have also added significant discussion about the AERONET retrievals to the new Section 2, including discussion about particle shape.
c) Although not specifically requested by the reviewer, we've also added some discussion about how the sC retrieval is altered if sC is 100% externally mixed (Section 5.2), and include this in our error analysis.

2 About Section 5 (Critique on Bond report)

I agree with the authors of the discussion paper that their approach is more consistent with the AERONET model and the measured radiance fields, and this has to be (and is already) stressed in the paper. A consistent approach like the one proposed is preferable to an inconsistent approach. However, I'm not sure about the importance of this consistency, as already the AERONET model applies a very strong (often unrealistic) assumption on the size-independence of the refractive index.

*Reviewer comments in **bold**.*

Yes, this is a very strong assumption that is usually unrealistic. It is even unrealistic to assume that all particles of any given size have the same refractive index. Other strong assumptions include the limited particle shapes (spheres and spheroids) and the homogeneous atmosphere used in the retrieval. Nonetheless, we argue that the AERONET model is the most realistic aerosol retrieval product available at this time.

We would also argue that this makes it even *more important* to maintain the link to the radiation field. That is, since the model is an oversimplified version of reality (like all models), it is very important to make sure that the results are consistent with the radiance and extinction measurements. Without that connection, the results don't necessarily have a connection to reality at all.

We have eliminated the old Section 5 and added a new Section 2 that provides additional details about the AERONET retrievals. Hopefully this clarifies the importance of the link to the radiance field.

My feeling is that "misconception" in the section title is too strong as the proposed methodology makes some assumptions (e.g. the scheme in Figure 6) that might also be called "misconceptions" from a strict physical point of view.

The "misconception" was meant to refer to the notion that somehow AAOD is a more reliable parameter than the parameters from which it is derived.

As I'm not really convinced that the Bond approach is so much worse at the end, I would suggest to call this section "Discussion of AAOD approach in Bond report" (or similar).

This is true -- the Bond approach might not be much worse in the end. We didn't mean to imply that the Bond approach was invalid; rather, we were trying to point out that our approach does not require more assumptions than the Bond approach (as stated in that report), and that there are some additional assumptions with the Bond approach that were overlooked in that report.

At any rate, we have abandoned this topic as a section. We have taken some of the material from this section, and incorporated it into Section 2 (Description of the AERONET retrievals).

3 Specific remarks
*** p13610 l3: 53% -> 50%**
*** p13612 l14 and l19: 0.700 m -> 0.7 m**
*** p13612 l19: insert absorption after hematite**
*** p13613 l1: it is unclear here which retrieval is meant, probably "our retrieval"**

We've made these changes. Thank-you.

*** p13613 l25: "... which implies that all particles are internally mixed." is the wrong conclusion, I think. I suggest "... have the same homogeneous refractive index. This implies that the refractive index from AERONET is some kind of effective refractive index."**

Reviewer comments in ***bold***.

We changed that passage to:

The operational AERONET product assumes that all particles have the same homogeneous refractive index, which implies that all particles are internally mixed (as in Figure 2b of Bond and Bergstrom, 2006).

We have also included the following paragraph in 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 here is our companion paper (Part 2), which is now available online. The important point here is that the AERONET retrieval model is an internal mixture, and not an external mixture. Sure, there are two types of internal mixing (homogeneous and heterogeneous), but both forms of internal mixing result in enhanced absorption of the sC particles. Additionally, both forms of internal mixing prevent the separation of sC AAOD from BrC AAOD with the AAE approach.

*** p13623 l3: "of carbon..."**

*** p13628 l15: If errors of size distribution and errors of refractive index compensate each other, the errors of derived parameters (AAOD, AAE, or SSA) could be smaller (I don't know if this is actually the case here). If the authors can not exclude that size and refractive index errors compensate each other or have further evidence, I suggest to remove the sentence in brackets.**

Interesting point. The text in question from the original manuscript reads:

"Once an optimal size distribution and refractive index are found, they are used to compute the AAOD, AAE, and other optical parameters reported in the AERONET database. (Thus, claims that AAOD, AAE, or SSA are somehow more robust than the retrieved refractive index are false, as all of these quantities are computed from the refractive index.)"

Perhaps we should have used the word "unsubstantiated" instead of "false," which would have been more accurate. We are unsure that changing a single word will clarify the sentence, though, so we have replaced this passage and moved it to Section 2. It now reads:

Once an optimal size distribution and refractive index are found, they are used to compute the AAOD, absorption Angstrom exponent (AAE), single-scatter albedo (ω_o), and other optical parameters reported in the AERONET database. Thus, all of the almucantar retrieval products are mathematically linked by Mie Theory and T-matrix theory, and we can not claim that one of *these parameters is more robust than another. The AERONET product is a “package" in this sense – taken together, all of the products provide a consistent set of parameters that produce the measured radiance field.*

The main point we are trying to emphasize is that AAOD, AAE, and SSA (and refractive index) are retrievals, not direct measurements. This affects how we can interpret the AERONET products, which we’ve discussed fairly extensively in the new Section 2 as well as Part 2 (now online).