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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Retrieval of aerosol composition directly from satellite and ground-based measurements" *by* Lei Li et al.

Anonymous Referee #2

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This paper describes a modification to the GRASP algorithm to generate retrievals of a predefined basis set of aerosol "species." Typically the GRASP and the Dubovik and King (hereafter DK) provide a retrieval of size and real/complex refractive index. This is then used by others to classify species. Here they tie the refractive indices retrieved by GRASP by a basis set aerosol species. Their primary point is that using a predefined basis set of species is more direct than inferring typing from the measured extinction and subsequently derived absorption angstrom exponent method (or in a few cases adding index of refraction) commonly used in the community. As I finished this review, I did do a quick comparison with reviewer 1. I would agree with their point that perhaps the most important aspect of this work is it provides something of a forward operator to perform more apples to apples comparisons between satellite and models to close

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the radiance fields. I also agree that discussion of this point should be expanded. This discussion needs to cover these points.

I think this effort moves the field forward in that they change the basis set as to what the retrieval is producing (BC, BrC, soluble and insoluble). However, these parameters are by no means equivalent to "composition," which is their premise for the entire paper. "Soluble and insoluble fractions" are wholly ill-defined in the context of composition, as they are related really to hygroscopicity. This is also true in part in regard to BC and BrC as they were functionally optical parameters long before we knew much of their true chemical nature. Here they are an indicator of spectral absorption properties. There are a myriad of soluble species with different indices of refraction and hygroscopic properties and likewise spectral dependencies of absorption based on mixtures. Thus, the idea that what is being retrieved as independent information on composition is fundamentally not true. What is a step forward, is they demonstrated that using the GRASP algorithm, you can generate a retrieval of parameters other than the standard size, index of refraction etc. as a basis of some categorization of the optical environment. One could look at this as a complex transform, but really it is simply a way of having an a priori set of basis functions for different aerosol species. But this is sort of what most traditional aerosol retrievals do, provide a best fit on the developers notion of what the aerosol environment looks like. Coarse mode dust, fine mode pollution, absorbing components, etc. So when the authors say they are the first ones to extract composition directly from satellite, this is not true either, it is just the first time in the GRASP algorithm has taken this approach. Rather, they are taking some liberty with the language. My first major comment is I think the authors need to be very careful about their language here. They should be more up front as to what they are doing, or spend much more time explaining why they think there is something fundamentally different in their approach. My second point is that it is unclear as to what is really going on in the retrieval. If one considers the GRASP algorithm simply has a smoothness of fit contain, but otherwise can pick any refractive index and size it wants, why not just take the standard GRASP algorithm as it is, and after the fact

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match the size and refractive index to any basis function they like? Or is that what they are already doing? (it is hard to tell in figure 1 and the associated discussion). Is the goodness of fit always the same from the standard GRASP algorithm, or does your predefinition of species leave a residual? If so, how big is that residual? They also list as an example the refractive index for ammonium nitrate, but what of the other species? We are referred to a GACP dataset, and Figure 3 has such dynamic range between species it is hard to tell even what these values are. I think an appendix needs to be generated that provides details on these key aspects of the retrieval. I think in order for them to prove validity of the algorithm, they should do the retrieval with their basis set, and then as a baseline compare to the standard grasp algorithm, and see to what extent the goodness of fit to the radiance fields changes.

My final major comment is that there is really very little verification work provide that shows that the results of the retrievals are fundamentally better than other categorization methods (that is getting back to the question in point 2 (if this method leaves a residual from the free running GRASP algorithm). Or can you baseline against a simple AE vs AAE plot for species? One could argue that real verification has always been an issue for retrievals. There are many studies that show that DK retrievals provide reasonable results. But, those studies and here pick sites that are generally single aerosol specie dominated (the once exception is Solar village). This is why at least a self-consistent baseline against the standard GRASP and DK retrievals is so important. The discussions in Section 4 related to Figure 14-20 global maps that provide some truthiness. But close examination (which required me zooming way into the plots) shows many logical inconsistencies, especially around coastlines, where the overall hydration of the particles leads to an increase in the “soluble fraction” Likewise there is a great deal of “insoluble” AOD in Brazil-even though we know smoke organic components do in fact have a hygroscopicity to them, even if it is low), as well as retrieval errors. Are they not really just applying a form Schuster’s “water fraction” algorithm? This then closes the loop with comment 1: are they really doing a soluble and insoluble aerosol specie? In the end, I appreciate what the authors are trying to do, and can see

how this will benefit the community. But there are logic issues that need to be wrung out, and some form of baseline verification that shows this method is actually taking us in the right direction.

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