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

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We appreciate the referee's time, constrictive and valuable comments that helped us to better consolidate and focus the manuscript. Our responses are presented below.

Anonymous Referee #2

General comments: 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 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. Response: We appreciate this very interesting review and valuable thoughts that pushed us to better communicate the novelty and originality of the proposed approach. First of all, we understand the importance and sensitivity to the semantic and agree that the word "composition" can be more associated with aerosol chemical composition, which does not properly reflect what is retrieved. However, to define as retrieval of aerosol type or classification will be wrong as well, as explained below. We therefore converged to aerosol "component" that will be used hereafter. We argue here and provide a discussion in the manuscript on why the suggested approach should be distinguished from the traditional aerosol retrievals (LUT approaches) and is fundamentally different to previous approaches. It should be noted that the retrieval of aerosol type has been clearly recognized as an important task by the scientific community and has been addressed in several studies. For example, there are a number of approaches that attempt to identify the type of aerosol through analysis of optical parameters such as single scattering albedo (SSA), Ångström Exponent (AE), AAE (absorption AE), refractive index, etc. Specifically, Russell et al. (2014) relate AERONET- and POLDER-derived optical properties to different aerosol types: urban, dust, marine, biomass burning, etc. Studies by Chung et al. (2010) and Bahadur et al. (2012) use AERONET optical properties like AE and AAE to separate BC, BrC, and dust into species-specific AAOT (absorption AOT). Schuster et al. (2005, 2009, 2016a) and Li et al. (2015) quantify the relative volume fractions of one or more aerosol species (e.g. BC, BrC, iron oxide, water) by adjusting the mixture of several components in an aerosol model to fit AERONET-retrieved refractive indices. However, our new approach differs substantially from all of these methods because it does not use a retrieval of optical parameters as an intermediate step. Thus, we expect the GRASP/Component approach to provide a stronger link to the radiation field than the previous approaches, as well as fundamentally higher retrieval accuracy. Moreover, some of above methods have additional differences and limitations compared to our proposed approach. For example, the Russell et al. (2014) approach is rather qualitative and does not attempt to quantify the relative volume or

mass fractions of different species in an aerosol mixture. Chung et al. (2012) and Bahadur et al. (2012) seem to use a technique for separating carbonaceous aerosols from dust that is not fully consistent with the AERONET retrieval assumptions, as discussed by Schuster et al. (2016b). Also, the Look-Up Table (LUT) approaches employed in most satellite retrievals (Martonchik et al., 1998; Remer et al., 2005; Kahn and Gaitley, 2015; Popp et al., 2016; Hammer et al., 2018; etc.) are designed to search amongst a preselected set of aerosol models (or their mixtures) for a model that provides the best fit to the observations. Since the models in a LUT are usually associated with a number of aerosol types (e.g. desert dust, smoke, urban aerosol etc.), the identification of the model that provides the best fit is often considered as a retrieval of aerosol type/composition. For observations with enhanced sensitivity, such as the Multi-angle Imaging SpectroRadiometer (MISR), a large number of models can be justified in the LUT and the differentiation of the models described by the ensembles of parameters can indeed be rather robust. However, LUT approaches are fundamentally limited to a discrete set of possible solutions, whereas the GRASP/Component approach searches through a continuous space of solutions; thus, the identification of aerosol components with our new methodology is significantly more detailed and elaborate. The proposed approach also bridges directly to the quantities of aerosol compositions used in the global chemical transport models. Specifically, our aerosol component retrievals can satisfy the requirements of chemical transport models to constrain their aerosol estimations on a large or global scale. However, we note that the GRASP/Component approach is only possible if 1) there is significant instrument sensitivity to the parameters that are related aerosol component (i.e. complex refractive index), and 2) this sensitivity is maintained while other parameters like the size distribution are adjusted. The retrieval provided in this article is unique because the aerosol component fractions are iterated in the original algorithm until a "best fit" is achieved with the measured radiance field, making it unnecessary to use retrieved optical properties as a constraint. Thus, the aerosol volume fractions determined with this new procedure have a direct link to the measured radiance field, whereas the procedures outlined above have a

direct link to the retrieved aerosol optical properties provided by AERONET or satellite products. In addition, the sensitivity of aerosol water fraction to the real refractive index is shown in Figure R1 (Schuster et al., 2009). Schuster et al. (2009) illustrated that the soluble aerosol components (sea salt, ammonium sulfate, ammonium nitrate, etc.) indicate similar refractive indices for similar mixing ratios, even though the dry refractive indices can be quite different. Hence, the aerosol water fraction can be derived from the mixture real refractive index if the aerosols are known to be one of the common soluble aerosols (in this study it is assumed as ammonium nitrate). Hygroscopicity, as an indicator of how aerosol growth in size or scattering, is related to relative humidity. However, in this study we are more concerned about how aerosol water content is related to real refractive index, which is much simpler (no hysteresis, no exponential growth, etc.) according to the study of Schuster et al. (2009) shown as Figure R1. These discussions are added to the manuscript.

Figure R1. Water fractions for two-component aerosol mixtures as a function of the real refractive index. (Schuster et al., 2009)

We also modified the abstract to better explain the novelty of the approach: "This study presents a novel methodology for remote monitoring of aerosol component over large spatial and temporal domains. The concept is realized within the GRASP (Generalized Retrieval of Aerosol and Surface Properties) algorithm to directly infer aerosol component from the measured radiances. The observed aerosols are assumed as mixtures of hydrated soluble particles embedded with black carbon, brown carbon, iron oxide, and other (non-absorbing) insoluble inclusions. The complex refractive indices of the dry components are fixed a priori (although the refractive index of the soluble host is allowed to vary with hydration), and the complex refractive index of the mixture is computed using mixing rules. The volume fractions of these components are derived together with the size distribution and the fraction of spherical particles, plus the spectral surface reflectance in cases the satellite data is inverted. The retrieval is implemented as a statistically optimized fit in a continuous space of solutions. This

contrasts with most conventional approaches where the type of aerosol is either associated with a pre-assumed aerosol model that is included in a set of Look-Up-Tables, or determined from the analysis of the retrieved aerosol optical parameters (e.g., single scattering albedo, refractive index, etc. provided by the AERONET retrieval algorithm); here, we retrieve the aerosol component explicitly. The approach also bridges directly to the quantities used in the global chemical transport models. We first tested the approach with synthetic data to estimate the uncertainty, and then applied it to real ground-based AERONET and space-borne POLDER/PARASOL observations; thus, the study presents a first attempt to derive aerosol component from satellites. Our results indicate aerosol optical characteristics that are highly consistent with standard products (e.g., R of $\sim$ 0.9 for aerosol optical thickness) and demonstrate an ability to separate intrinsic optical properties of fine- and coarse-sized aerosols. We applied our method to POLDER/PARASOL radiances on the global scale and obtained spatial and temporal patterns of the aerosol component that agree well with the knowledge on aerosol sources and transport features. Finally, we discuss limitations and perspectives of this new technique. This approach is different from the conventional methods that use post-processing of the retrieved aerosol optical properties for aerosol typing. The proposed method assumes observed aerosols as mixtures of particles composed of black carbon, brown carbon, absorbing insoluble, non-absorbing insoluble embedded in a soluble host. The algorithm then derives size distribution and the fractions of these components. The complex refractive index of each component is fixed a priori and the complex refractive index of mixture is computed using mixing rules. The approach is first tested with synthetic data and the uncertainties are estimated. Then, it is applied to the real ground-based AERONET and space-borne POLDER/PARASOL observations, known to be sensitive to aerosol complex refractive index. The study presents a first attempt to derive aerosol component from satellites. The obtained aerosol optical characteristics are highly consistent with the standard products (R of $\sim$ 0.9 for aerosol optical thickness). The approach also presented an ability to separate between aerosol properties in fine and coarse size fractions, in case of POLDER/PARASOL and

AERONET. Examples of application to POLDER/PARASOL on the global scale are presented. The obtained spatial and temporal patterns of the aerosol component agree well with our knowledge on aerosol sources and transport features. Finally, limitations and perspectives are discussed."

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Kaufman, Y., Tanré, D., Mattoo, S., Chu, D., Martins, J., Li, R., Ichoku, C., Levy, R., Kleidman, R., Eck, T., Vermote, E., and Holben, B.: The MODIS aerosol algorithm, products and validation, Journal of the Atmospheric Sciences, 62, 947-973, 2005. Russell, P., Kacenelenbogen, M., Livingston, J., Hasekamp, O., Burton, S., Schuster, G., Johnson, M., Knobelspiesse, K., Redemann, J., Ramachandran, S., and Holben, B.: A multiparameter aerosol classification method and its application to retrievals from spaceborne polarimetry, J. Geophys. Res., 119, https://doi.org/10.1002/2013JD021411, 2014. Schuster, G.L., Dubovik, O., Arola, A.: Remote sensing of soot carbon – Part 1: Distinguishing different absorbing aerosol species, Atmos. Chem. Phys., 16, 1565–1585, https://doi.org/10.5194/acp-16-1565-2016, 2016a. Schuster, G.L., Dubovik, O., Arola, A., Eck, T.F., Holben, B.N.: Remote sensing of soot carbon – Part 2: Understanding the absorption Ångström exponent, Atmos. Chem. Phys., 16, 1587–1602, https://doi.org/10.5194/acp-16-1587-2016, 2016b. Schuster, G.L., Dubovik, O., Holben, B.N., Clothiaux, E.E.: Inferring black carbon content and specific absorption from Aerosol Robotic Network (AERONET) aerosol retrievals, J. Geophys. Res., 110, D10S17, https://doi.org/10.1029/2004JD004548, 2005. Schuster, G.L., Lin, B., Dubovik, O.: Remote sensing of aerosol water uptake, Geophys. Res. Lett., 36, https://doi.org/10.1029/2008GL036576, 2009.

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 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. Response: Regarding the goodness of fit and validity of the new algorithm as compared to the standard GRASP algorithm: we thank for bringing up this point because it is indeed an essential step that was done in early stages of the new algorithm developments, but is not explicitly mentioned in the original manuscript. Figure 11 of the original version, however, already showed an inter-comparison of correlations between optical characteristics derived by the new approach, the standard GRASP and the operational AERONET product, which addresses in a way the raised question. Here, in Figure R2, we present the residuals from different GRASP approaches. The data subset used is the same as for the inter-comparison with the operational AERONET product (Figure 11 and Table 4). Figure R2 shows that the residuals of the GRASP/Component approaches (Maxwell-Garnett (MG) and volume weighted (VW) mixing rules) are almost the same as those of standard GRASP. Although the average residual of aerosol component approach (2.4±0.9% for MG and 2.4±1.0% for VW) is slightly higher than of standard GRASP (2.3±0.9%), the maximum residual of aerosol component approach (5.0% for MG and 5.7% for VW) is somewhat smaller than that of standard GRASP (6.6%). The next sentence is added to the manuscript: "In addition, the GRASP/Component approach produces almost the same average residual (2.4±0.9% for MG and 2.4±1.0% for VW) as that of the standard GRASP algorithm (2.3±0.9%) while the maximum residual for GRASP/Component (5.0% for MG and 5.7% for VW) is smaller than that for standard GRASP (6.6%); ± denotes standard deviation."

Figure R2. Inter-comparison of retrieval residual among different GRASP approaches. Red color represents the Maxwell-Garnett (MG) mixing model; blue - the volume-weighted (VW) mixing model; and black - the standard (ST) GRASP/PARASOL product that do not employ the aerosol component retrievals. The data subset used is the same as for the inter-comparison with the operational AERONET product (Table 4 and Figure

11 of the manuscript).

Regarding the used refractive indices: while addressing the first specific comment of the first reviewer, we added a table summarizing description of aerosol components and refractive indices (Table 2). In addition, an axis break is added for the ordinate in Figure 5a to better show the different range of real refractive indices. We also modified Figure 1 in the manuscript (see Figure R3 below).

Figure R3. General structure of GRASP algorithm with aerosol component conversion model, courtesy of (Dubovik et al., 2011). The red dashed frames represent modifications for the component inversion approach. fˆ* represents vector of inverted measurements, aˆP represents vector of unknowns at the p-th iteration, ãĂŰf(aãĂŮˆP) represents vector of measurement fit at the p-th iteration.

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. Response: The first part of this comment is indeed getting back to the question in point 2. Figure R2 and the discussion related to Figure 11 of the manuscript reply to this concern by showing the equivalence of the residuals and consistency of the retrievals between the new and the standard approaches. Regarding global maps, due to overall high complexity of the aimed approach, it is possible that not everywhere the retrieved aerosol components fit perfectly the expectations and a better understanding of what the retrieval represent is desirable. At the same time, not fully logical results are likely caused by a physical reason such as lack of sensitivity to some specific aerosol component, since optical observations in some cases sensitive to certain optical equivalent. This is challenge not only for the proposed methodology and for any other effort aimed to derive aerosol composition from remote sensing observations. In this respect, is can be noted that many known features are correctly represented in our retrievals. Moreover, some not yet known or fully understood phenomena can also be responsible for some "anomalies" and the proposed data set can be very useful basis for formulating new scientific question. Nevertheless, we agree that a more detailed and zoomed analysis should be in scope of future studies. However, due to very large volume of produced data in this publication and limited resources, we made the choice to present an overview of the results. Finally, we believe that the provided above and in the corrected version of the manuscript clarifications explain better the originality of the approach and the sense of the derived aerosol components.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-208/acp-2019-208-AC2-supplement.pdf

C11

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-208, 2019.
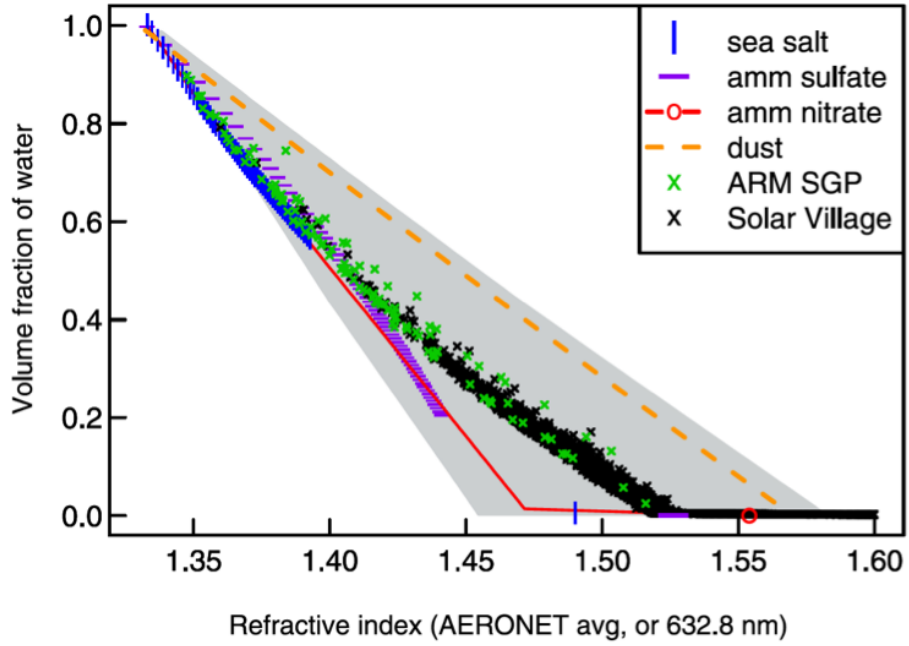
Fig. 1. Figure R1. Water fractions for two-component aerosol mixtures as a function of the real refractive index. (Schuster et al., 2009)
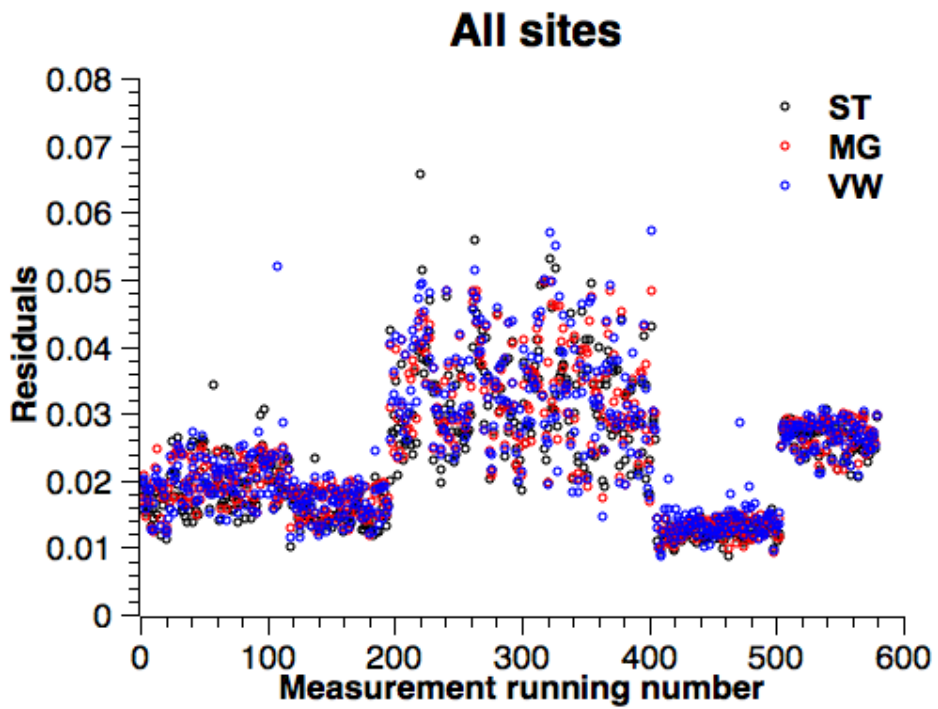
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Fig. 2. Figure R2. Inter-comparison of retrieval residual among different GRASP approaches.

C14

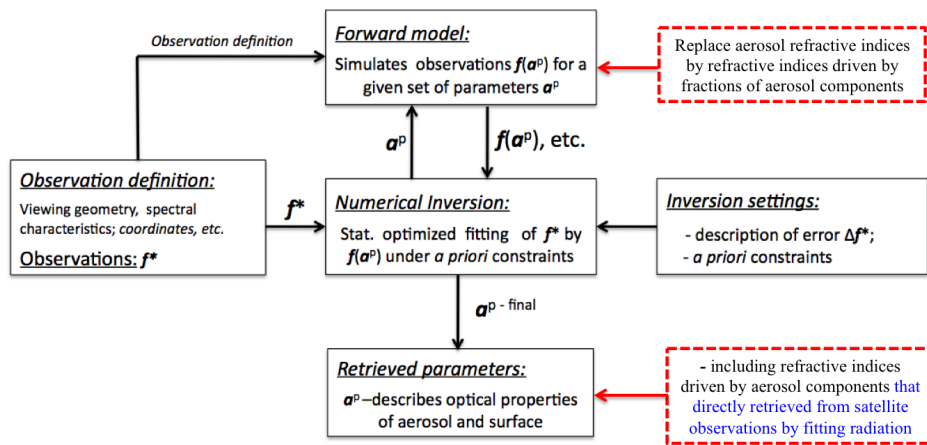## General structure of inversion algorithm

Fig. 3. Figure R3. General structure of GRASP algorithm with aerosol component conversion model, courtesy of (Dubovik et al., 2011).

C15