Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-208-AC1, 2019

Atmospheric Chemistry and Physics
Discussions
Open Access

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

Lei Li et al.

lilei@radi.ac.cn

Received and published: 19 July 2019

Response to comments of referee #1 on “Retrieval of aerosol composition directly from satellite and ground-based measurements” by Lei Li et al.

We appreciate the referee’s thoughtful reading, valuable suggestions and time that we hope helped us to improve the manuscript. Our point-to-point replies are presented below.

Anonymous Referee #1 This paper presents an interesting strategy for using polarized/multiangular satellite measurements to infer aerosol composition. The method uses the Generalized Retrieval of Aerosol and Surface Properties (GRASP) along with assuming that aerosols are mixtures of non-soluble particles embedded within a sol-

Printer-friendly version

Discussion paper

uble host. GRASP then derives size distribution, loading and light absorption characteristics based on determining the fractions of each aerosol type. The algorithm is applied first on synthetic data, then on ground-based AERONET, then on historic POLDER data.

General comments: Although quite long, I find this paper to be well-organized, well-written and worthy of publication. I really like the idea that the GRASP retrieval can combine specific aerosol types (size + shape + absorption) to retrieve the aggregate. This makes it possible to compare directly with models (that also assume these types). I think the authors should highlight this even more than they have already. Finally, this is probably well beyond the scope of the paper, But I would like to see some sort of graphic (even for a single panels from Figs 15-19), to compare how your global maps (of species type) compare with other maps, such as from AeroCom models, and/or MISR size/shape/absorption climatology. Right now, they look reasonable, but I would be curious whether they might “change” our way of thinking about aerosol types distribution. Response: We appreciate the referee’s suggestion. The direct bridging to the models is highlighted now in the abstract and the discussion is extended in the manuscript. Also, in fact, a comparison of our GRASP retrievals with GEOS-5/GOCART model estimations was already done. However, as the referee mentioned, this next step is beyond the scope of the current publication. Either this observation-based aerosol components maps will “change” or not our way of thinking about aerosol types distribution is highly interesting and desires a more focused effort. Nevertheless, to give a glance on how the retrievals agree with the models, a couple of additional figures are presented below. Figure R1 shows the correlations for monthly averaged dust mass concentrations over the globe for 2008. It shows, for example that the global average coarse dust mass concentration retrieved by GRASP (125.5 mg/m2) is lower than that simulated by GEOS-5/GOCART (174.7 mg/m2). Figure R2 shows the map of coarse mode dust distribution in April 2008. The retrievals tend to provide lower coarse dust concentration over the mineral dust region (such as in Northern Africa and Northern China), which is consistent with the previous studies showing that models overestimate mineral dust in dust-dominated places (Jones and Christoper, 2007; Ginoux et al., 2001, Chin et al., 2009). Also, the observations show quite a fine spatial distribution structure.

Figure R1. Correlation between the monthly averaged dust mass concentrations of GRASP retrievals and GEOS-5/GOCART model estimations over the globe in 2008.

Figure R2. Distribution of dust mass concentration estimated by GEOS-5/GOCART model and retrieved by GRASP/POLDER in April 2008.

Chin, M., Diehl, T., Dubovik, O., Eck, T. F., Holben, B. N., Sinyuk, A., and Streets, D. G.: Light absorption by pollution, dust, and biomass burning aerosols: a global model study and evaluation with AERONET measurements, Ann. Geophys., 27, 3439-3464, 2009. Ginoux, P. Chin, M., Tegen, I., Prospero, J. M., Holben, B., Dubovik, O., and Lin, S.-J.: Sources and distributions of dust aerosols simulated with the GOCART model, 106(D17), 20255-20273, 2001. Jones, T. A. and Christopher, S. A: MODIS derived fine mode fraction characteristics of marine, dust, and anthropogenic aerosols over the ocean, constrained by GOCART, MOPITT, and TOMS, J. Geophys. Res., 112(D22), 2007.

Specific comments: some small suggestions for improving the paper. 1) Specifically, what are the aerosol type/model components? I feel as if a table could be used to describe each component, its size/shape and refractive index components. Response: This useful suggestion is addressed by presenting a table summarizing the aerosol components description. The complex refractive indices of each component are presented at 0.440 ðİIJĞðİŚŽ and 0.865 ðİIJĞðİŚŽ.

2) Around line 290, the terms Frac(i) and dV/dlnr appear without definition. I guess they are in the table 1, but since table 1 (in the PDF) wasn't near the text, one might want to define first time in the text. Response: Thank you for noticing this. We added the definitions of “Frac(i) and (dV(r_j))/dlnr (i=1,. . .,N_r)” in the text.

3) The stars on equation (1) are confusing. Do the stars represent a priori or solutions? Response: The term with a star represents satellite measurements. For example, f_iˆ* denotes a vector of the measurements and f_i denotes a vector of the estimations. It is now clarified in the text.

4) In line 355, Csph appears (see comment #2). Response: The C_(sph) definition was added in the text.

5) How are the intrinsic aerosol parameters allowed to vary in time and space? Response: Besides the aerosol component retrieval module, the presented algorithmic developments essentially rely on the available heritage of the previous retrieval developments for POLDER space instrument. Thus, as in the standard GRASP algorithm (Dubovik et al., 2011), the satellite retrieval is designed as a statistically optimized simultaneous fitting of the observations over a group of pixels implemented under additional inter-pixel constraints. Specifically, the variations of the retrieved parameters horizontally from pixel-to-pixel or temporary from day-to-day over the same pixel are limited by the additional a priori constraints, in a similar manner to how it is applied in inverse modeling by Dubovik et al. (2008). The inclusion of these additional constraints is expected to provide retrieval of higher consistency for aerosol retrievals from satellites, because the retrieval over each single pixel will be benefiting from coincident aerosol information from neighboring pixels, in addition to the information about surface reflectance (over land) obtained in preceding and consequent observations over the same pixel.

Dubovik, O., Herman, M., Holdak, A., Lapyonok, T., Tanré, D., Deuzé, J.L., Ducos, F., Sinyuk, A., Lopatin, A.: Statistically optimized inversion algorithm for enhanced retrieval of aerosol properties from spectral multi-angle polarimetric satellite observations, Atmos. Meas. Tech., 4, 975–1018, https://doi.org/10.5194/amt-4-975-2011, 2011. Dubovik, O., Lapyonok, T., Kaufman, Y.J., Chin, M., Ginoux, P., Kahn, R.A., Sinyuk, A.: Retrieving global aerosol sources from satellites using inverse modeling, Atmos. Chem. Phys., 8, 209–250, https://doi.org/10.5194/acp-8-209-2008, 2008.

6) For Eq (7), there appear to be a lot of zeros in the matrix. We can assume there is no covariance? For example, I can't see avc (volume size distribution) and av (total volume) as being independent Response: Zeros in the matrix denote that there are no smoothness constraints for these parameters, such as the spectral smoothness constraint. A clarification sentence is added now in the text.

7) Line 396-397: I guess I am curious, what do you mean by: "choice of mixing rule. . .significantly affects the results". Can you show something about this? Response: This statement should be indeed clarified. The meaning is that the different assumptions on mixing rules, which are often used for estimation of aerosol composition, were reported by Xie et al. (2014) as affecting the aerosol composition retrieved by an independent algorithm that uses ground-based AERONET measurements. For instance, the Bruggeman approximation was found as more suitable for the dust case, the Maxwell-Garnett for the haze case, and volume average for the clean case. In our study, the Maxwell-Garnett and volume average mixing rules were employed independently of the aerosol type and the retrieval results are inter-compared (Figure 11). In our approach we have not identified a significant influence of the mixing rule choice on the quality of the retrievals. Moreover, the aerosol optical properties were rather well comparable in both cases. The fractions of the elements present some differences due to the differences in the formulation, but are still in a reasonable agreement. The text is modified accordingly.

Xie Y., Li Z., Li L. et al.: Study on influence of different mixing rules on the aerosol components retrieval from ground-based remote sensing measurements. Atmospheric Research, 145: 267-278, 2014.

8) It appears that Eq (11) and Eq (12) have the same RHS? Response: No, please note that there is a difference in the sign before ε_r.

9) Line 448 looks like a formula (11 minus 12) not 11 and 12. Response: It is corrected.

10) Lines 455-484: See my comment #1. Response: A table summarizing description

of the aerosol components is added (see Table 2 in the revised manuscript and the response to comment #1).

11) I have a few comments regarding figure 5 and paper text. Could Fig 5a be split into two? There is a huge range of real refractive indices for CAI, but not for BrC and NAI. I cannot tell if the differences in assumptions for BrC and NAI around 1.5 are significant. Response: An axis break is added for the ordinate in Figure 5a to better show the different range of real refractive indices.

12) Line 532-540. These range of values could also be added to a table (e.g. #1) Response: The values of complex refractive indices at 0.440 ðİIJĞðİŚŽ and 0.865 ðİIJĞðİŚŽ, used in the uncertainty test, are added to Table 2 (Please see the response to comment #1).

13) Line 545-546: “Elevated” meaning larger loadings or higher altitudes? Response: It is reworded to “Therefore, the estimates should be quite reasonable in the cases of large pollution loading.”

14) Lines 547-550: Are the fractions of BC and CAI somehow constrained so they can’t be “large”? I note that they never approach 0.5 and barely approach 0.1 Response: It is important to say that the maximal retrieved fractions for BC and CAI (mainly representing iron oxides) do not result from a constrain, despite have a limit due to the range of possible complex refractive indices in the pre-computed kernels of aerosol optical characteristics. That is, the volume fractions of these two highly absorbing species are indeed limited in the algorithm to 10% for BC and 3% for CAI, based on the below listed reasons. However, our analysis showed that these maximal values were never reached in the inversion procedure. Previous in-situ studies (Ganor and Foner, 1996; Guieu et al., 2002; Lafon et al., 2004, 2006, Alfaro et al., 2004; Wagner et al., 2012; Formenti et al., 2014) showed that iron oxides account for 2.8âĂŤ6.5% of mineral dust by mass. It means approximately 1.4âĂŤ3.25% by volume, since the density of free iron is much higher than other common minerals (4.28 g cm-3 for goethite and 5.25 g

cm-3 for hematite, as opposed to 2.65 g cm-3 for illite, kaolinite, quartz, and calcite; Formenti et al., 2014 The fraction of BC in atmospheric aerosol was generally reported not exceeding 10% (Bond et al., 2013). It is also to note that the retrievals of aerosol composition derived from AERONET measurements by Schuster et al. (2016) demonstrated that the volume fraction of free iron remains relatively constant in West Africa throughout the year (1.4âĂŤ1.7%) and the volume fraction of black carbon reaches a peak of 1.0% for the fine mode during West African biomass burning season and a peak of 3.0% for the fine mode in southern Africa biomass burning. The corresponding precision is added to the manuscript (lines 590 to 609).

Alfaro, S., Lafon, S., Rajot, J., Formenti, P., Gaudichet, A., and Maille, M.: Iron oxides and light absorption by pure desert dust: an experimental study, J. Geophys. Res., 109, D08208, doi:10.1029/2003JD004374, 2004. Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., Deangelo, B.J., Flanner, M.G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P.K., Sarofim, M.C., Schultz, M.G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P.K., Jacobson, M.Z., Kaiser, J.W., Klimont, Z., Lohmann, U., Schwarz, J.P., Shindell, D., Storelvmo, T., Warren, S.G., Zender, C.S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380–5552, https://doi.org/10.1002/jgrd.50171, 2013. Formenti, P., Caquineau, S., Chevaillier, S., Klaver, A., Desboeufs, K., Rajot, J.L., Belin, S., Briois, V.: Dominance of goethite over hematite in iron oxides of mineral dust from Western Africa: Quantitative partitioning by X-ray absorption spectroscopy, J. Geophys. Res. Atmos., 119, 12740-12754, https://doi.org/10.1002/2014JD021668, 2014. Ganor, E. and Foner, H. A.: The mineralogical and chemical properties and the behavior of aeolian Saharan dust over Israel, in: The Impact of Desert Dust Across the Mediterranean, edited by: Guerzoni, S., and Chester, R., Kluwer Academic Publishers, Printed in the Netherlands, 163-172, 1996. Guieu, C., Loye-Pilot, M. D., Ridame, C., Thomas, C.: Chemical characterization of the Saharan dust end-member: Some biogeochemical implications for the western Mediterranean Sea, J. Geophys. Res., 107 (D15), 4258,

Printer-friendly version

Discussion paper

https://doi.org/4210.1029/2001JD000582, 2002. Lafon, S., Rajot, J. L., Alfaro, S. C., Gaudichet, A.: Quantification of iron oxides in desert aerosol, Atmos. Environ., 38, 1211-1218, 2004. Lafon, S., Sokolik, I.N., Rajot, J.L., Caquincau, S., Gaudichet, A.: Characterization of iron oxides in mineral dust aerosols: Implications for light absorption, J. Geophys. Res. Atmos., 111, 1–19, https://doi.org/10.1029/2005JD007016, 2006. 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, 2016. Wagner, R., Ajtai, T., Kandler, K., Lieke, K., Linke, C., Müller, T., Schnaiter, M., and Vragel, M.: Complex refractive indices of Saharan dust samples at visible and near UV wavelengths: a laboratory study, Atmos. Chem. Phys., 12, 2491–2512, doi:10.5194/acp-12-2491-2012, 2012.

15) For the plots of Figure 6, I am wondering what the "uncertainty" is. Should I read this as uncertainty is fraction (%) of fraction? What if these were presented in same units as x-axis (fraction)? Of course estimates of tiny fractions should have large % uncertainties, (but then that also means that the estimates of the fraction of the other elements will have lower % uncertainties). Response: Thank you for this comment, it is indeed not sufficiently explained in the text. The uncertainty is defined in percentage as (retrieved_fraction-assumed_fraction)/assumed_fraction. We would prefer to leave the notation in Figure 6 as it is, however, an explanation is added in the paragraph on uncertainty calculation (lines 646 to 647).

16) Line 581. I don't understand: "The non-absorbing insoluble can stand also for the insoluble organic carbon" Response: The sentence is reworded. The intention was to be mentioned in the text problematic that the non-absorbing dust and non-absorbing organic carbon have similarity in the complex refractive index and is not distinguishable for the employed type of observations. It is corrected to "The non-absorbing insoluble component can represent not only non-absorbing dust, but also non-absorbing organic carbon, as was mentioned above."

17) Some of the figures have panels with cut-off axes (e.g. Fig 3) Response: Corrected.

Printer-friendly version

Discussion paper

18) Are the units Fig 15-19 correct? (mm3/m2)? Response: It can be confusing, but the units are correct. The units denote the volume concentration in total atmospheric column with unit surface area. We clarified it in the text.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-208/acp-2019-208-AC1-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-208, 2019.

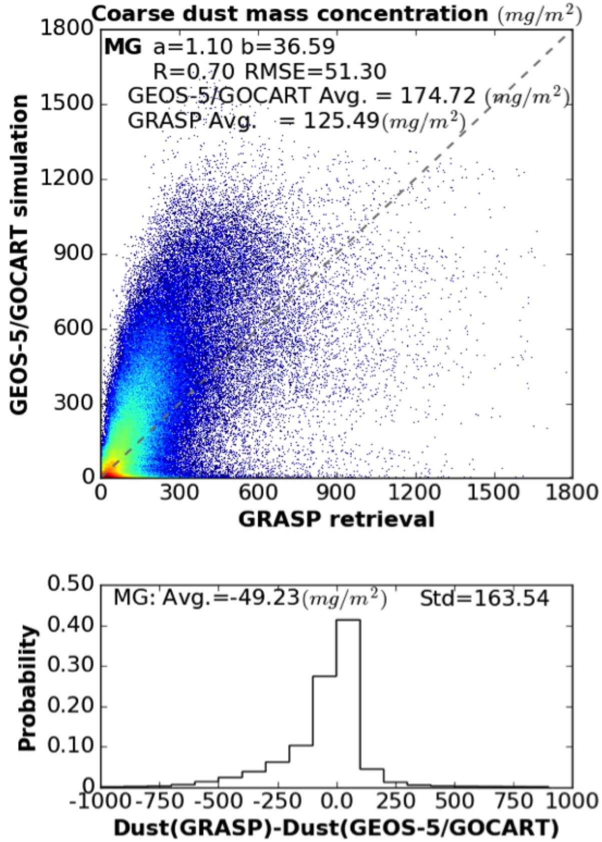

Fig. 1. Figure R1. Correlation between the monthly averaged dust mass concentrations of GRASP retrievals and GEOS-5/GOCART model estimations over the globe in 2008.

Printer-friendly version

Discussion paper

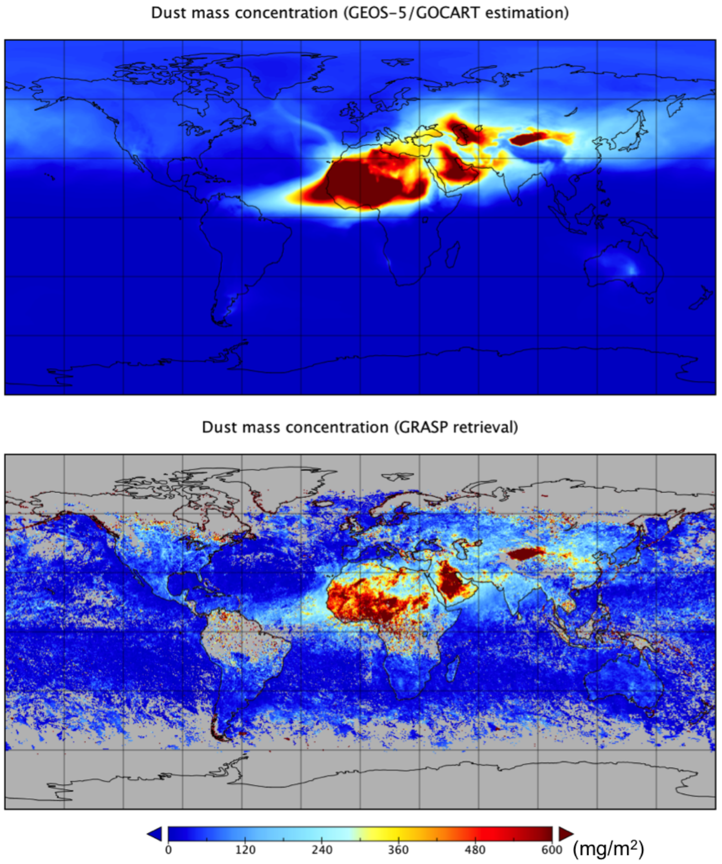

Fig. 2. Figure R2. Distribution of dust mass concentration estimated by GEOS-5/GOCART model and retrieved by GRASP/POLDER in April 2008.

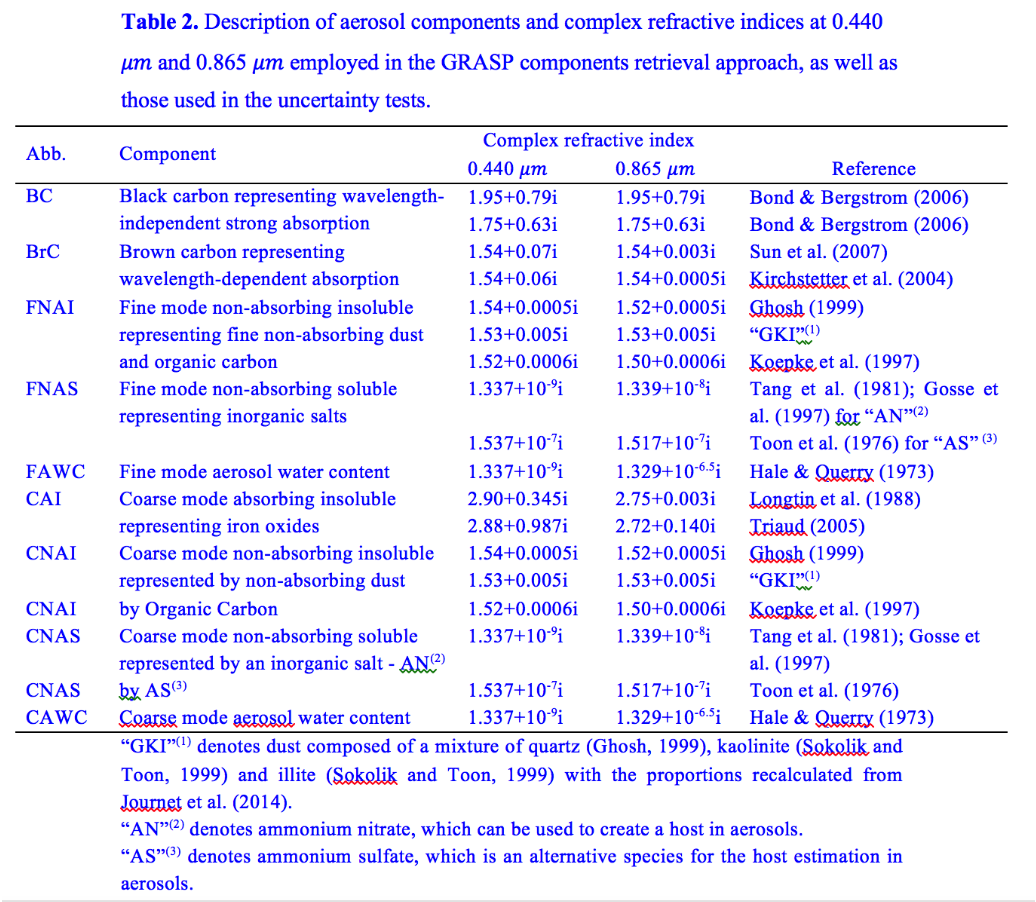

Table 2. Description of aerosol components and complex refractive indices at 0.440 *μm* and 0.865 *μm* employed in the GRASP components retrieval approach, as well as those used in the uncertainty tests.

Abb.	Component	Complex refractive index		Reference
		0.440 *μm*	0.865 *μm*	
BC	Black carbon representing wavelength-	1.95+0.79i	1.95+0.79i	Bond & Bergstrom (2006)
	independent strong absorption	1.75+0.63i	1.75+0.63i	Bond & Bergstrom (2006)
BrC	Brown carbon representing	1.54+0.07i	1.54+0.003i	Sun et al. (2007)
	wavelength-dependent absorption	1.54+0.06i	1.54+0.0005i	Kirchstetter et al. (2004)
FNAI	Fine mode non-absorbing insoluble	1.54+0.0005i	1.52+0.0005i	Ghosh (1999)
	representing fine non-absorbing dust	1.53+0.005i	1.53+0.005i	"GKI"(1)
	and organic carbon	1.52+0.0006i	1.50+0.0006i	Koepke et al. (1997)
FNAS	Fine mode non-absorbing soluble representing inorganic salts	$1.337+10^{-9}i$	$1.339+10^{-8}i$	Tang et al. (1981); Gosse et al. (1997) for "AN"(2)
		$1.537+10^{-7}i$	$1.517+10^{-7}i$	Toon et al. (1976) for "AS" (3)
FAWC	Fine mode aerosol water content	$1.337+10^{-9}i$	$1.329+10^{-6.5}i$	Hale & Querry (1973)
CAI	Coarse mode absorbing insoluble	2.90+0.345i	2.75+0.003i	Longtin et al. (1988)
	representing iron oxides	2.88+0.987i	2.72+0.140i	Triaud (2005)
CNAI	Coarse mode non-absorbing insoluble	1.54+0.0005i	1.52+0.0005i	Ghosh (1999)
	represented by non-absorbing dust	1.53+0.005i	1.53+0.005i	"GKI"(1)
CNAI	by Organic Carbon	1.52+0.0006i	1.50+0.0006i	Koepke et al. (1997)
CNAS	Coarse mode non-absorbing soluble represented by an inorganic salt - AN(2)	$1.337+10^{-9}i$	$1.339+10^{-8}i$	Tang et al. (1981); Gosse et al. (1997)
CNAS	by AS(3)	$1.537+10^{-7}i$	$1.517+10^{-7}i$	Toon et al. (1976)
CAWC	Coarse mode aerosol water content	$1.337+10^{-9}i$	$1.329+10^{-6.5}i$	Hale & Querry (1973)

"GKI"(1) denotes dust composed of a mixture of quartz (Ghosh, 1999), kaolinite (Sokolik and Toon, 1999) and illite (Sokolik and Toon, 1999) with the proportions recalculated from Journet et al. (2014).

"AN"(2) denotes ammonium nitrate, which can be used to create a host in aerosols.

"AS"(3) denotes ammonium sulfate, which is an alternative species for the host estimation in aerosols.

Fig. 3. Table 2. Description of aerosol components and complex refractive indices at 0.440 ðİIJĞðİŠŽ and 0.865 ðİIJĞðİŠŽ employed in the GRASP components retrieval approach, as well as those used in the uncertainty tests.