Referee report to the "The Aerosol Limb Imager: Multi-spectral Polarimetric Observations of Stratospheric Aerosol" manuscript by Daniel Letros et al.

The manuscript describes a retrieval algorithm for the new Aerosol Limb Imager (ALI) instrument and presents some results from the synthetic retrievals and from three example measurements made by the instrument. Although the measurement concept of ALI is similar to that used by the upcoming ALTIUS mission of ESA, it offers an unique feature of measuring the polarization state of the limb-scatter radiance. Authors did a great job showing how this feature of ALI can be used to detect contamination by clouds, which has always been an issue for limb-scatter aerosol retrievals. Less impressive are, however, the results from the aerosol retrieval itself. Here, I got an impression that the polarizationsensitive measurements make the instrument useless for the aerosol retrieval. This is not an issue for the scientific significance of the paper but the authors, especially PIs of the project, should think about if they really want to provide this impression to the scientific community. Unfortunately, the presentation of the results is elaborated quite poor and needs to be improved. To improve the message of the paper authors need to quantify the required conditions for the retrieval of reasonable aerosol extinction coefficient profiles from real measurements. A careful proof-read of the paper is needed to correct typos, extra or missing words etc.

General comments

- Date and time of the ALI measurements and those of the collocated reference measurements are not provided. This make impossible to understand which aerosol conditions were investigated. Furthermore, no information about collocation criteria is given.
- Simulation results do not look representative enough. The aerosol extinction above 30 km was set to zero and no attempts were made to check what happens if a realistic aerosol distribution at high altitudes is included. It is not clear for which surface albedo the simulations are done and what happens if albedo changes. How the estimations of the surface albedo are affected by the presence of the aerosol above 30 km? Are the simulation results remain the same if an aerosol profile for different aerosol loading conditions is used? Logarithmic plots and absence of relative difference plots make evaluation of the retrieval quality nearly impossible. If I understand it correctly, the synthetic retrievals were done without adding measurement noise to the simulated spectra. How well the retrieval chain works if measurement noise is added (I mean here using noisy simulated spectra, not only adding the noise covariance matrix into the retrieval)? How the aerosol parameters for the exercise with bimodal aerosol PSD were set, where comes the information about the used parameters from? Are they realistic? Some quantification of the results in the case of the bimodal PSD is needed, i.e. some realistic bimodal distributions corresponding to different aerosol loading conditions have to be found and used for simulations. Comparison of the

- mode radius and width from the unimodal and bimodal distributions does not make much sense. A comparison of the effective radii might be more useful.
- Comparison with measurement data is difficult to evaluate without having the relative difference plots. It is not clear for which aerosol conditions the comparison with reference data was made, this information is extremely difficult to derive from the logarithmic plots. The overall conclusion from the comparison seems to be that only one of the three measurements produces reasonable results although data from both OSIRIS and OMPS-LP work well in all three cases. Is it a general issue of the applied technique? Is the technique then useful at all? A more detailed discussion of the usability of ALI results needs to be done at this point.

Detailed comments

- Line 6 and 34: At this point it is unclear what the term "scalar width" means.
- Introduction lacks some information about how the ALI instrument is ranged with respect to the previous, present and planned space-borne aerosol instruments.
- Section 2: Some technical data of the instrument need to be provided, e.g. vertical/horizontal resolution and sampling, FOV range, typical exposure time etc.
- Lines 51 52: "The SPS also contains one linear polarizer after the LCR and another after the AOTF to further refine the polarized image" There is a polarizer before the AOTF and AOTF itself passes one polarization direction through. Please explain shortly why an additional refinement is needed.
- Figure 1 is quite dark and depending on the monitor and illumination difficult to see. Please light up the dark parts of the figure.
- Figure 2, panel (b): the abbreviation "DE" is not defined in the caption.
- Line 69: "The spectral bandpass of ALI for a tuned frequency is taken as the full-width half-max (FWHM) of the diffraction response." it looks like, here, you use the "diffraction response" term to describe the same function as shown in the panel (b) of Fig. 2 an referred to as the "spectral response". In line 75 you write, however "spectral response is calculated by integrating the measured diffraction responses" which suggest different meaning of these two terms. Please clarify.
- Figure 3 caption: "LCR on state" here and throughout the text, it would be less confusing is you wrote "on" in the quotation marks. The same is for "off".
- Line 99: please give some details to explain what the "calibrated broadband integrating sphere of known (randomly polarized) spectrum" is.

- Line 100: "Furthermore, since they reproduce the spatially flat and full-field conditions of the integrating sphere, they can be used to relate the ALI measurements to the external source each pixel measured."— Does "spatially flat" means homogeneous of a flat shape of the source? It is not quite clear how to get a response to an inhomogeneous source signal if you have a response to a homogeneous full-field signal. Please explain. Maybe I misinterpret what you want to say with this sentence, in this case please rephrase for more clarity.
- Line 128: Are there any investigations of the temporal stability of the obtained calibration parameters and possible dependence on the ambient temperature and pressure, strength of the illumination etc. ?
- Figure 3 caption: "White dots in this image indicate the bad pixels as determined by the dark correction." I do not see any white dots in panel (a), what do the black dots mean?
- Figure 5 caption: The sentence "The radiance profiles of (b, c, e, f, h, i) are constructed by following Section 2.3 to convert images into photons/s/cm2 /sr from DN. Then following Section 2.1 to obtain units of photons/s/cm2 /sr/nm." should be moved to the main text.
- Figure 5 caption: "the solar irradiance produced by a radiative transfer forward model" solar irradiance is not produced by radiative transfer models.
- Line 206: "a regularization matrix in place of S_a^{-1} " S_a^{-1} is also a regularization matrix, do you mean Twomey-Tikhonov regularization matrix, i.e. a smoothing constraint here?
- Line 208: "measurement and state vectors of large dynamic range which tends to produce ill-conditioned inversions." the ill conditioning of the inverse problem is not caused by the large dynamic range of the measurement and state vectors. Its reasons are rather a dense vertical sampling, insensitivity of the retrieval to certain vertical ranges and correlation between different parameters.
- Line 214 217: In most previous aerosol retrievals the albedo and aerosol retrieval were run alternatively within an iterative process allowing the retrieved albedo to adjust to the retrieved aerosol profile and vise versa. In this retrieval, the albedo is retrieved only once before the aerosol retrieval. Additional investigations need to be done to show that the retrieved albedo does not depend significantly on the a priori aerosol profile used for the albedo retrieval.
- Line 221 222: I do not expect that the influence of aerosol at 33 34 km is minimal. This statement has to be proven using measurement data, e.g. GloSSAC.
- Sect. 4.1 please provide the aerosol profile used for albedo retrievals. Dependence of the albedo retrieval on the assumed aerosol profile needs to be investigated.

- Lines 245 246: "Therefore, we also present an approach to retrieve the Stokes parameters of the atmosphere in the Stokes basis of ALI ..." Please explain what "the Stokes basis of ALI" means.
- Eq. (7): What happened to U and V components. Were they just neglected? Please clarify.
- Lines 267: "...which directly correspond to tangent altitudes at the time the observation was taken" What do you want to highlight with "at the time the observation was taken", are the tangent heights measured not at the same time?
- Lines 300 302: "In this example Θ received little to no action by the inversion to adjust it form the a-priori state. It is sensible to simply not include Θ as a property in x and just rely on the a-priori values in the forward modeling." Is it a common behavior or does it just happen occasionally?
- Line 310: "... we convolve the smoothed DoP with a central difference impulse response." please explain what is the "a central difference impulse response" or give a reference.
- Fig. 7: Please provide an illustration what happens if there is a strong aerosol level with larger particles in place of the cloud layer.
- Line 333: "At each scan, the true state of the atmosphere includes GloSSAC aerosol but unlike the exercise of Fig. 7 and Fig. 8 no ice layer is included." please show the aerosol profile used in the retrieval (if you like in the Supplement) and prove the results are the same for a different aerosol loading scenario.
- Sect. 4.2.4: It would be interesting to have a larger statistics about the working and non-working DoP retrievals.
- Line 349: "retrieved along side a scalar width" you still haven't properly defined what scalar width means.
- Line 369: "as the finest resolution A produced" What does "A" mean here?
- Lines 368 370: Resolution might be limited by the forward model grid but not determined by it. It is mainly determined by the instrument FOV and sampling and also applied regularization. What you mean here is the sampling.
- Line 371: "we do not employ regularization" Using S_a is also a regularization.
- Lines 391 394: The description looks like the Levenberg-Marquard method, why do not you use the name, is the any significant difference?
- Sect. 4.3.1: Please show the GloSSAC aerosol profile used for the study. Why was only one profile used? Dependence on the true aerosol profile might be significant. Different aerosol loading conditions need to be investigated.

- Line 400: Please provide the illustration what happens if you do not set the aerosol profile above 30 km to zero.
- Figure 10: Is it correct that the true surface albedo is not changed here?
- Line 416: What happens if a priori and true widths are the same but the width is included into the retrieval?
- Figs. 10 13, 15, and 16 must include relative differences.
- Line 433: "We wish to emphasize that the limb measurements of ALI are polarized, and we speculate that this polarized content contains useful information about the aerosol phase scattering matrices which is of course influenced by the particle sizes" also non-polarized measurements contain information on the particle sizes.
- Sect. 4.3.1: Authors found a strong dependence of the results on the presence of the second mode for highly polarized measurements, the question is if polarized measurements are really an advantage or a drawback. This should be discussed in more details in the text.
- Lines 448 451: "With respect to the simulations done in the exercise surrounding Fig. 9, we find almost all behavior regarding Fig. 14 to be expected including: the relative balance between horizontal and vertical polarizations for all three scans, the spectral regions expected to fail given the polarimetric response of ALI in Scan 1 and Scan 3, and the relative magnitude of the DoP for Scan 1 and Scan 2." this discussion is difficult to follow, please write in more details what exactly you expect from the discussion around Fig. 9 and what we see in this respect in Fig. 14.
- Lines 448 451: "The simulation of this geometry done for Fig. 9 produced a true state DoP approximately ranging between 0.3 0.4" please provide corresponding plots, if you prefer they can be in the Supplement.
- Line 473: "... of interest is the profile of r which indicates a layer of larger particles at approximately 22.5 km..." please compare this results with SAGE III data and make a statement about the agreement.
- Lines 485 488: A large disagreement for ALI and a good agreement for other instruments means, in my opinion, that the instrument concept where the polarized measurements are used is not optimal for the retrieval of the aerosol extinction coefficient. Please discuss this issue.

Technical corrections

• Line 33 and throughout the text: "dependant" \longrightarrow "dependent"

- Figure 1 caption: "comprises of one off-axis" \longrightarrow "comprises one off-axis"
- Figure 2 caption: "shown in black" \longrightarrow "is shown in black"
- Figure 3 caption: "response of ALI (Letros et al., 2024) shown as" \longrightarrow "response of ALI (Letros et al., 2024) is shown as"
- Line 117: "be used to constructed" \longrightarrow " be used to construct"
- Line 128: "conversion to from DN into" \longrightarrow "conversion from DN into"
- Line 170: "Here we discuse the concepts" \longrightarrow "Here we discus the concepts"
- Line 175: "and marks the lower altitude limit to retrieve" \longrightarrow "and mark the lower altitude limit to retrieve"
- \bullet Line 347: "to emphasise that the in the context" \longrightarrow "to emphasise that in the context"
- Line 347: "criteria we set is to retrieve" \longrightarrow "criterion we set is to retrieve"
- Line 349: "their is" \longrightarrow "There is"
- \bullet Line 364: "ceiling on of the radiance" \longrightarrow "ceiling on the radiance"
- Line 396: "summary of retrieval result" "summary of retrieval results"
- Line 407: "by retrieving a N and r profile" —> "by retrieving N and r profiles"
- Line 413: "by the biased caused" do you mean "by the biases caused"?
- Line 417: "shows the behaviour" \longrightarrow "show the behaviour"
- Line 515: "but we this is a point" \longrightarrow "but this is a point"