Response to Reviewer 2 comments

General comment: This is a review of the manuscript titled “Vertical Profiles of Aerosol Chemical Species Concentration retrieved through Synergy of Spaceborne Lidar and Polarimeter Observations” submitted to AMT by Merdji et al.

The paper describes an analysis of the performance of the AEROCHEMPro algorithm to retrieve aerosol properties from combined lidar and polarimeter data, based on simulated observations. The subject is very relevant as the combination of lidar and polarimeter is very promising to advance aerosol observations. The paper is generally well written and figures are clear. However, there are many limitations and assumptions that need to be explained and highlighted more before the paper can be accepted. Hence major revisions are needed as detailed below.

Response: The authors thank the reviewer for the positive assessment of the manuscript and for recognizing the relevance of the AEROCHEMPro framework in the context of advanced lidar-polarimeter synergy. We appreciate the constructive feedback regarding the limitations and assumptions inherent in the current synthetic evaluation.

We agree that a clearer and more critical discussion of these assumptions is essential for providing a realistic assessment of the algorithm’s performance. In the revised manuscript, we have significantly expanded the discussion of limitations and assumptions. The specific points raised by the reviewer have been addressed individually in the detailed responses below, and the manuscript has been updated to provide a more balanced perspective on the current capabilities and future challenges of the AEROCHEMPro methodology.

Comment 1: Limitations leading to optimistic results:

There are many limitations and simplifications in the work and analysis that need to be explained and highlighted more. I understand many assumptions are due to practical limitations of the analysis and some is left to future work. However, they need to be clearly stated in the manuscript and in the abstract and conclusions to inform the reader of the scope of the paper. Especially the implications of these simplifications need to be made clear, as they may mostly lead to overly optimistic results. Limitations that I identified are listed below (limitations 1 to 5).

Response: We thank the reviewer for this constructive overview. We completely agree that a clear, transparent discussion of our study's limitations, simplifications, and their potential to lead to optimistic results is vital to properly inform the reader of the paper's scope. In line with the reviewer's recommendations, we have carefully revised the Abstract, Methodology, and Conclusions sections to explicitly state these underlying assumptions and highlight their implications. We address each of the specific limitations identified by the reviewer in the individual responses below, detailing exactly how the manuscript has been updated to balance the presentation of our performance results.

Comment 1/Limitation 1: If I understand correctly, the forward model and assumptions for the simulated observations and the retrievals are entirely consistent. Hence they make the same simplifications. If I am correct, please make this explictely clear. This will result in a overly optimistic result, which then also need to be discussed.

Response: Clarification added. Thank you for the comment. The reviewer is correct that the forward model and inversion assumptions are entirely consistent. We acknowledge that this "internal consistency" creates a best-case scenario that does not yet account for "forward model errors" (e.g., discrepancies between real-world particle shapes and the theoretical models used in the retrieval).

In Section 2.3 (Methodology): We added a sentence stating: "*The ensemble of aerosol properties retrieved by AEROCHEMPro, which are the same variables used in the forward model to simulate measurements, is presented in Table 5.* ***It is important to note that the forward model used to generate pseudo-observations and the inversion framework are fully consistent. This setup is intended to establish the theoretical baseline performance of AEROCHEMPro before introducing the further complexities of real-world instrument-to-model mismatches. Thus, the current results correspond to a best-case scenario.***" (Page 20, lines 489- 493)

In the Conclusion: We updated the text to highlight that these results represent the "theoretical lower bound of uncertainty leading to overly optimistic results. "*This study presents the innovative AEROCHEMPro retrieval approach, designed to retrieve vertical profiles of aerosol chemical species and their optical properties across diverse atmospheric conditions and regions. Detailed case study analysis and statistical comparisons based on a simulated atmosphere across multiple transects at a global scale reveal both its principal strengths, its unprecedented capabilities, and some limitations of the approach.* ***In particular, because this study relies on a self-consistent simulation framework over a spatially uniform surface baseline, these results represent an idealized theoretical performance limit and a lower bound on retrieval uncertainty.*** *Performing single-pixel retrievals over a homogeneous surface minimizes complex surface-aerosol signal mixing, presenting an optimistic baseline. Future validation phases will incorporate cross-model testing and spatially heterogeneous terrain configurations to evaluate AEROCHEMPro against real-world surface discrepancies rigorously.*" (Page 45, line 867-875)

Abstract Text Update: "As this initial validation relies on a self-consistent simulation framework over a spatially uniform surface baseline, these results represent an optimistic, idealized theoretical performance limit." (Page 2, line 40-42)

Comment 1/Limitation 2: The surface parameters are not fitted according to section 2.1.6. Partitioning between aerosol and surface signals is one of the main challenges in aerosol remote sensing, so this is a very important limitation again leading to overly optimistic results.

Response: Clarified and explanation added. We thank the reviewer for highlighting this critical aspect of aerosol remote sensing. The partitioning between aerosol and surface signals is indeed a fundamental challenge that can significantly influence retrieval accuracy.

We clarify an important point: Within the AEROCHEMPro methodology, the land and ocean surface reflection parameters are jointly retrieved within the retrieval state vector rather than being mathematically omitted or hard-coded as fixed constants.

However, the reviewer is entirely correct that our experimental setup represents an idealized, optimistic scenario. This optimism stems from the physical design of the simulation scene, where **the underlying true surface properties were kept uniform across the simulated transect**. Within our single-pixel inversion configuration, the land BRDF and water surface models utilize standard, loose smoothness constraints (Lagrange multipliers of 10^{-4} to 10^{-3}), leaving them free to adapt. Only the polarized surface component uses a tight constraint (10.0) to stabilize its spectral behavior.

Because the algorithm successfully recovers these parameters close to their uniform baseline values with limited surface parameter interference, it establishes a clean theoretical performance baseline for the AEROCHEMPro aerosol speciation logic. We fully acknowledge this limitation and have thoroughly revised the text in Section 2.1.6 and Section 4 to ensure absolute transparency.

Revised Manuscript Text Update (Section 2.1.6): "*While GRASP allows joint retrieval of surface and aerosol properties (Dubovik et al., 2021), we adopted a constrained single-pixel retrieval strategy for this baseline evaluation. Surface parameters were jointly adjusted in the state vector. On the other hand, they were initialized with spatially uniform values across the transects and subject to strong a priori constraints (Lagrange multipliers), thus significantly limiting their spatial variability during retrievals. Specifically, a high weight was assigned to the spatial variability constraint of the polarized surface component and the BRDF kernels to maintain stability (See Table 6 for details). This configuration effectively minimizes pixel-to-pixel surface variability, allowing the inversion to focus on the vertical aerosol speciation without significant interference from surface-aerosol signal mixing. We acknowledge that this approach represents an idealized scenario; while it establishes a theoretical performance baseline for AEROCHEMPro, future work will involve more flexible surface-atmosphere retrievals to explicitly capture the complexities of heterogeneous terrain.*" (Page 12, line 295-304)

Revised Manuscript Text Update (Section 4 - Conclusions): "*In particular, because this study relies on a self-consistent simulation framework over a spatially uniform surface baseline, these* *results represent an idealized theoretical performance limit and a lower bound on retrieval uncertainty. Performing single-pixel retrievals over a homogeneous surface minimizes complex surface-aerosol signal mixing, presenting an optimistic baseline. Future validation phases will incorporate cross-model testing and spatially heterogeneous terrain configurations to evaluate AEROCHEMPro against real-world surface discrepancies rigorously.*" (Page 45, line 870-875)

Comment 1/Limitation 3: From the very low differences between simulated and retrieved U/I in table X, I suspect that the geometries used for the observations and retrievals are all assuming the principal plane, in which U is essentially zero. Polarimetric retrievals are sensitive to geometry and a principal plane geometry may be considered a best-case scenario. Please make clear which geometry is used and discuss the implications of the simplifications in this assumption (if applicable).

Response: Clarification added. The reviewer is correct: the simulated observations and subsequent retrievals were performed assuming a principal plane geometry. In this configuration, the Stokes U component is essentially zero, which provides an idealized polarimetric signal focused on the Q component's sensitivity to aerosol microphysics. We chose this geometry to establish a theoretical performance baseline for the lidar-polarimeter synergy without the added complexity of azimuthal dependencies. We agree that this represents a "best-case scenario." In the revised manuscript, we have made this assumption explicit in Section 2.2 and have removed the U/I residuals from the results analysis and Figure 7, as they do not provide a meaningful assessment of the retrieval's performance in this geometry. We have added a discussion acknowledging that more realistic off-principal plane geometries will introduce a non-zero U component, which will be addressed in future sensitivity studies.

1. Update to Methodology (Section 2.2): "*In this study, all synthetic observations were calculated within the principal plane geometry. In this configuration, the solar and viewing vectors are co-planar, and the Stokes U component is essentially zero. This provides an idealized polarimetric signal, allowing us to isolate the sensitivity of the Q component to aerosol microphysics and chemical speciation. While this represents a 'best-case scenario' for polarimetric retrieval, it serves as a necessary baseline for evaluating the vertical profiling capability of the AEROCHEMPro framework.*" (Page 12, line 317-321)

2. Update to Results discussion (Figure 7):

"*Figure 7 illustrates the agreement between measured and fitted polarimeter observations for all pixels and spectral channels across the selected transect, providing a direct verification of how the residuals relate to the injected noise levels. For the intensity component (I), the mean biases are negligible, with normalized absolute values ranging from 3.03×10^{-5} to $3.56\ 10^{-4}$, and standard deviations ranging between 1.28×10^{-3} and 2.97×10^{-3}. These standard deviations align with the relative 3% noise floor applied to the typical fractional normalized radiance levels of* *the scene. The normalized polarization residuals (Q/I) show minimal mean biases ranging from 3.62×10^{-5} to 1.22×10^{-3}, with standard deviations remaining stable across wavelengths, hovering around 0.003 (from 2.71×10^{-3} to 3.30×10^{-3}). This specification falls within the absolute noise floor of 0.005 assigned to the underlying Stokes parameters for the AOS polarimeter. As the simulations are performed in the principal plane, the U component is excluded from the analysis, as it does not carry independent information in this geometry. The consistency of the method is confirmed by the absence of systematic biases across the eight spectral bands."* (Page 30, line 639-648)

Comment 1/Limitation 4: The first guess used in the inversion is very important, as a first guess too far away from the truth may result in a non-convergence, while using the truth as first guess may lead to overly optimistic results. The first guess is not discussed in the manuscript, neither is a definition of (non-)convergence. Please discuss this and also the implication if a overly optimistic first guess is used (if applicable).

Response: Clarification added. We agree that a robust retrieval must be independent of its initialization. In the revised manuscript (Section 2.4), we have clarified our initialization strategy and provided a formal definition of convergence:

Initial Guess: To avoid "overly optimistic" results, we used a "cold-start" initialization across all global pixels. These values were intentionally kept generic and independent of the "true" pseudo-reality. As detailed in Table 6, this configuration is completely generic and independent of the "true" pseudo-reality across all retrieved state vector parameters:

- Chemical Composition Fractions: Aerosol chemistry is initialized using fixed, generic volume fractions across the modes, providing the solver with no prior geographic knowledge of the actual aerosol species.
- Vertical Profiles: All aerosol modes are initialized with a completely flat, spatially uniform vertical distribution (0.01 m^{-1} across all 100 layers), forcing the algorithm to retrieve the layer heights entirely from the observational constraints.

Convergence Definition: Convergence was strictly defined by a stopping threshold of 1.0×10^{-9} for the relative change in the cost function within a logarithmic minimization framework. The inversion is allowed up to 15 Levenberg-Marquardt iterations per pixel. We have updated Section 2.4 to clarify this formulation and explicitly state vector initialization.

"The minimization of Eq. (14) is performed using the Levenberg-Marquardt algorithm with a logarithmic convention to ensure the physical positivity of retrieved quantities. To ensure a robust global minimum is reached and to completely avoid dependence on the initial state, the state vector is initialized globally using a generic, spatially uniform 'cold-start' configuration. This includes fixed initial chemical species volume fractions and a flat vertical profile distribution across all layers as detailed in Table 6, ensuring the initialization is sufficiently

generic and does not favor specific solutions. We apply a stringent convergence threshold of 1.0×10^{-9} *for the relative change in the cost function, with a maximum of 15 iterations per pixel. Furthermore, the Jacobian matrix is computed with a finite difference scale of* 5.0×10^{-5} *to precisely capture the sensitivities between the aerosol species concentrations and the multifaceted observations."*(Page 26, line 579-587)

Table 6. *Summary of AEROCHEMPro/GRASP Single-Pixel Retrieval Parameters, Initialization (first guess), Bounds, and Smoothness Regularization.*

State Vector Variable	Physical Meaning	Initialization (first guess) & Bounds	Smoothness Regularization (Order of finite difference & Lagrange parameter (γ))	Notes
$C_{vol,size}$	Normalized concentrations of size bins corresponding to the aerosol size distributions (dV(r)/dlnr in 3 modes, 7 bins)	Init: (volume fraction per bin): fine mode = (0.015, 0.07, 0.015); dust (coarse) mode = (0.02, 0.08); sea-salt (coarse) mode = (0.02, 0.08). Bounds for each bin: 5×10^{-6} — 5.0.	Fine mode: 2nd order, $\gamma = 5\times10^{-3}$ Dust & Sea salt mode: 1st order, $\gamma = 5\times10^{-5}$	Fixed radius & σ_g per bin (Sec. 2.1.6)
$Frac^F{}_i$ $Frac^D{}_i$ $Frac^{SS}{}_i$	Aerosol chemical composition (volume fractions of 8 chemical species per mode)	Init: 0.01–0.4; Bounds: 10^{-6} – 0.95	No constraint (S=0)	Retrieves $Frac_i$ instead of the refractive index.
$Prof_{mode}$	Vertical distribution – 3 modes	Uniform init: 0.01 (100 layers)	No constraint (S=0)	Prevents sharp, unrealistic gradients.
C_{sph}	Particle sphericity fraction	Init: 0.999 (fine, sea salt); 0.01 (dust)	No constraint (S=0)	Aerosol mode shape
BRDF	Land surface reflectance modeled with Ross–Li BRDF kernels (isotropic + volumetric + geometric terms)	Parameter 1: value=[0.1]; min=[0.001]; max=[1.2] (λ_1–λ_8) Parameter 2: value=[0.1]; min=[0.01]; max=[2.0] Parameter 3: value=[0.1]; min=[0.01]; max=[1.0]	Parameter 1: 1st-order diff., $\gamma=1.0\times10^{-4}$ Parameter 2–3: 0th-order diff., $\gamma=0.0$	Core BRDF model for land; parameter 1 describing isotropic reflection is spectrally dependent, 2–3 volumetric and geometric are wavelength-independent
BPDF	Polarized bidirectional reflectance over land using Maignan–Breon formulation	value=[2.1]; min=[0.01]; max=[15.03] (λ_1–λ_8)	Parameter 1: 1st-order diff., $\gamma=1.0\times10^{1}$	Polarized land surface reflectance term
COX	Water surface bidirectional reflectance using Cox–Munk isotropic model	Parameter 1: value=[0.01–0.005]; min=[0.0001]; max=[0.05] (λ_1–λ_8) Parameter 2: value=[0.9]; min=[0.8]; max=[1.0] Parameter 3: value=[0.01];	Parameter 1: 1st-order diff., $\gamma=1.0\times10^{-3}$ Parameter 2–3: 0th-order diff., $\gamma=0.0$	Parameter 1 controls slope variance (wind speed), parameter 2 Fresnel term, parameter 3 foam fraction

min=[0.0015]; max=[0.1]

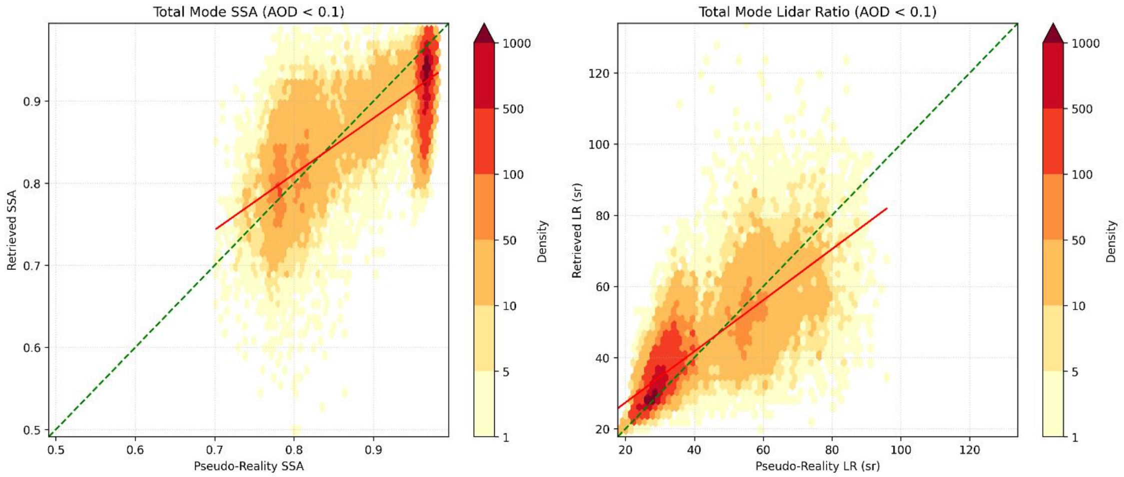
Comment 1/Limitation 5: In line 732 it is mentioned that only cases with AOD>0.1 are considered. Most of the global AOD values are below 0.1, so this is is also a limitation that need to be highlighted more.

Response: We thank the reviewer for highlighting this limitation. To address this concern, we performed a granular pixel-tracking analysis on the excluded low-AOD data (AOD < 0.1). We discovered that the systematic underestimation of SSA under clean conditions is not a global feature, but a clearly localized artifact confined to the High Southern Latitudes (> 60º S).

- Out of the 26,221 low-AOD pixels where the true atmosphere exhibits high scattering properties (SSA > 0.95), more than 95% (25,107 pixels) are located exclusively over the southern polar region. In this domain, the combination of an ultra-clean atmosphere (AOD < 0.1) and highly restricted, unoptimized satellite viewing geometries creates an exceptionally low atmospheric signal-to-noise ratio (SNR), mathematically degrading the retrieved SSA (Figure A).
- Global Recovery Outside the Polar Region: Once these high-latitude pixels are excluded, the systematic bias for the rest of the globe completely disappears. The Total SSA Mean Bias Error (MBE) drops to virtually zero (+0.0006 for AOD = 0.08-0.10 and -0.0012 for AOD = 0.06-0.08) (Figure B).

These statistics show that outside the polar region, the framework performs robustly even at low AOD. However, to protect the global dataset from these severe, unphysical high-latitude artifacts, the operational AOD > 0.1 threshold is required as a vital quality control mask. We have revised Section 3.2 to include these statistics and clarify this geographic context.

Manuscript Text Update (Section 3.2): "*The lower limit acts as a vital quality control mask; under near-pristine conditions (AOD < 0.1), the atmospheric signal-to-noise ratio degrades significantly, particularly for intensive properties like SSA at high southern latitudes (above* 60^{o} *S). The upper limit avoids extremely high aerosol loadings where strong multiple scattering and saturation effects can degrade retrieval accuracy.*" (Page 39, line 766-769)



Fgure A. Intensive properties (total column SSA and LR) at the South Pole region (> 60º S) only for all the transects.

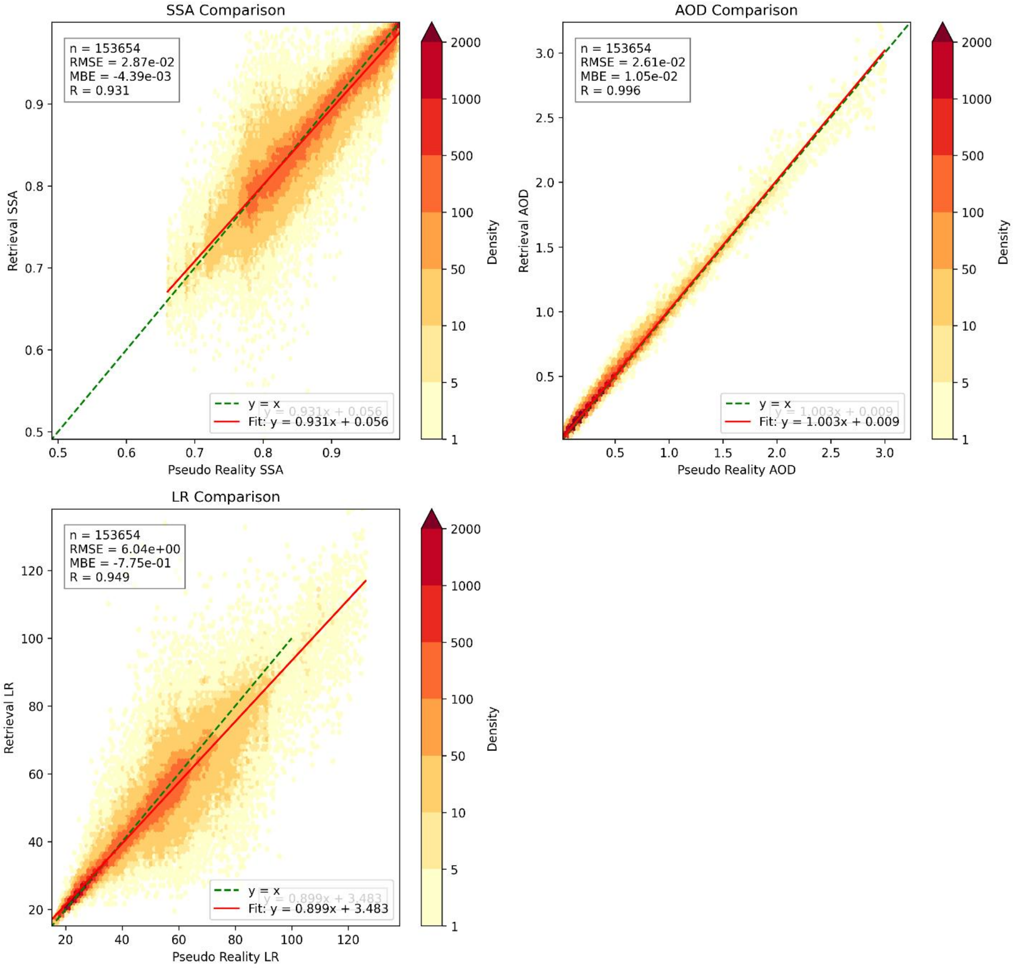


Figure B. Intensive properties excluding the South Pole region (> 60º S) for all the transects.

Comment 2: Polarimeter noise assumptions:

Generally multi-angle polarimeters aim for an absolute accuracy of the degree of linear polarization (DoLP) better than 0.005, as also indicated for the AOS-MAP in line 456. In their error model, the authors add 3% noise to I and 0.5% noise to Q and U. Depending on the specific values of I, Q, U and DoLP, this can lead to noise greater or smaller than 0.005 on DoLP. Please discuss in the paper the relation between the choices made for the applied noise and the target of DoLP absolute uncertainties being below 0.005.

Response: Clarification added. We thank the reviewer for highlighting this and catching the confusion. The reviewer is entirely correct to question this: stating that we applied a 0.5% relative noise to Q and U was a typographical error in our original manuscript.

Our simulation framework actually applies a strict absolute standard deviation noise floor of 0.005 ($\sigma_Q = \sigma_U = 0.005$) directly to the polarized Stokes parameters Q and U, while applying a 3% relative noise floor ($\sigma_I = 0.03 \times I$) to the total intensity I.

Using standard first-order Gaussian error propagation, the total absolute uncertainty (standard deviation) propagated onto the Degree of Linear Polarization ($DoLP = \frac{\sqrt{Q^2+U^2}}{I}$) is expressed as:

$$\sigma_{DoPL} \sqrt{\left(\frac{0.005}{I}\right)^2 + (DoLP \times 0.03)^2}$$

This mathematical relationship demonstrates that our error model represents a deliberately conservative stress test for the AEROCHEMPro algorithm across all typical scenarios, rather than an overly optimistic one:

- In Bright Scenes ($I > 1$): While a higher intensity suppresses the standalone polarimetric noise component below 0.005 ($I < 0.005$), the addition of the 3% intensity noise component maintains a total propagated error σ_{DoLP} of approximately sim 0.007, which strictly exceeds the 0.005 target threshold.
- In Dark Scenes ($I < 1$): As the normalized intensity drops, the polarimetric error term inflates rapidly. For instance, at $I = 0.5$, the propagated error component from Q and U doubles to 0.010, pushing the total uncertainty σ_{DoLP} to 0.011—more than twice as harsh as the mission specification.

Because our total simulated noise model matches or heavily exceeds the baseline instrument specifications in all viewing geometries, we ensure our retrieval validation remains highly robust. We have thoroughly revised Section 3.3 to remove the typographical error and make this conservative mathematical implementation explicit.

“*To evaluate the inversion performance against these mission requirements within our self-consistent simulation framework, the synthetic error model applies a 3% relative noise floor to* *the total intensity (I) and an absolute noise floor of 0.005 directly to the linear polarized Stokes parameters (Q and U). This absolute configuration directly governs the target uncertainty of the mathematically derived DoLP. For bright viewing geometries where the normalized intensity baseline scales above unity (I > 1), the absolute contribution of the standalone polarimetric components is suppressed, yet the combined total error on DoLP remains robust due to the simultaneous propagation of the intensity noise. Conversely, under low-signal conditions where I < 1, the framework allows the simulated noise to dynamically exceed the 0.005 specification. This configuration provides a conservative signal-to-noise magnitude of the retrieval logic, guaranteeing that the framework evaluates and proves the stability of AEROCHEMPro under realistic instrument performance constraints"* (Page 20, line 470-479)

Other specific comments:

Comment 3: Table 3: The number of significant digits varies throughout the table. Please make this consistent.

Response: Corrected. Thank you for pointing this out. We have revised Table 3 to ensure that the number of significant digits (and decimal places) is strictly consistent across all columns and parameters.

Comment 4: Table 4: There are additional entries for Nadir resolution and Swath width in the table. I assume these are threshold values. Please make this clear.

Response: Clarification added. Thank you for this comment. The reviewer is correct; these entries represent the mission design requirements (thresholds) compared to the expected operational performance. We have clarified this distinction in the updated Table 4 by restructuring the entry under "Geometric performance constraints" as follows:

Geometric performance constraints:

- Nadir spatial resolution:

- *Requirement (design threshold): 0.5 km*
- *Expected/actual performance: 1 km*

- Swath width:

- *Requirement (design threshold): 300 km*

Expected/actual performance: 100 km

Comment 5: Line 500: Please do not use math mode for the text in the equations

Response: Corrected. Thank you for this formatting correction. We have revised Line 500 and reviewed all equations throughout the manuscript to ensure that standard text, multi-letter abbreviations, and units inside math mode are properly formatted

Comment 6: Section 2.3.1: Here the surface parameters are described as part of the retrievals set up. However, they are not retrieved according to section 2.1.6. It is unclear how the surface parameters are kept fixed. Is that through the gamma assumptions? Please explain clearly in the text that the surface parameters are fixed and how this is achieved.

Response: Clarification added. We thank the reviewer for the opportunity to clarify this distinction. The surface parameters are indeed jointly adjusted and retrieved within the state vector, as shown in Section 2.3.1 and Table 5, rather than being numerically locked or omitted from the inversion loop.

When we note that the surface parameters are "fixed," we are referring to they are spatially uniform. It is not a constraint hardcoded into the inversion algorithm. Across the simulated transect, the true underlying surface properties were kept entirely uniform from pixel to pixel when generating the pseudo-observations.

The Surface reflectance updated text is as follows: *"While GRASP allows joint retrieval of surface and aerosol properties (Dubovik et al., 2021), we adopted a constrained single-pixel retrieval strategy for this baseline evaluation. Surface parameters were jointly adjusted in the state vector. On the other hand, they were initialized with spatially uniform values across the transects and subject to strong a priori constraints (Lagrange multipliers), thus significantly limiting their spatial variability during retrievals. Specifically, a high weight was assigned to the spatial variability constraint of the polarized surface component and the BRDF kernels to maintain stability (See Table 6 for details). This configuration effectively minimizes pixel-to-pixel surface variability, allowing the inversion to focus on the vertical aerosol speciation without significant interference from surface-aerosol signal mixing. We acknowledge that this approach represents an idealized scenario; while it establishes a theoretical performance baseline for AEROCHEMPro, future work will involve more flexible surface-atmosphere retrievals to explicitly capture the complexities of heterogeneous terrain."* (Page 12, line 295-304)

Comment 7: Line 610 and further: Instead of "10-5" use E or superscript.

Response: Clarified. Thank you for this suggestion. We have updated Line 610 and all subsequent occurrences throughout the manuscript to use proper scientific superscript notation (e.g., 10^{-5}) instead of the hyphenated text format.

Comment 8: Figure 7:

- How do these differences relate to the added noise levels? Explain in the manuscript
- As discussed in the main comments I suspect that the geometries are assumed to be principal plane, in which U is zero. It then is also meaningless to report on the errors on U, so then I suggest to leave this out and explain in the text.

Response: Clarification done. We thank the reviewer for this insightful comment. In an internally consistent closed-loop simulation framework free of forward model mismatch errors, the measurement residuals (Measured - Fitted) are driven purely by the random noise injected into the pseudo-observations. The standard deviations (std) computed across all subplots in Figure 7 explicitly demonstrate this relationship, proving that the single-pixel inversion successfully fits the observations precisely down to their statistical noise floor without introducing systemic biases.

Specifically:

- For Normalized Radiance (I, Left Column): The residual standard deviations range between 1.28×10^{-3} and 2.97×10^{-3}. Because the injected noise is a relative 3% of the normalized radiance, these values align perfectly with typical fractional baseline radiance levels across the track (e.g., a normalized radiance between 0.05 and 0.10 naturally yields an expected 3% noise standard deviation between 0.0015 and 0.0030).
- For the Normalized Polarization State (Q/I, Right Column): The residual standard deviations are remarkably stable and consistent across all wavelengths, hovering around 0.003 (ranging from 2.71×10^{-3} to 3.30×10^{-3}). This spread falls safely within the strict absolute standard deviation noise floor of 0.005 applied to the underlying Stokes components, confirming optimal convergence.

We have thoroughly updated Section 3.4 to include this quantitative noise comparison.

Manuscript Text Update (Section 3.4):

"Figure 7 illustrates the agreement between measured and fitted polarimeter observations for all pixels and spectral channels across the selected transect, providing a direct verification of how the residuals relate to the injected noise levels. For the intensity component (I), the mean biases are negligible, with normalized absolute values ranging from 3.03×10^{-5} to $3.56\ 10^{-4}$, and standard deviations ranging between 1.28×10^{-3} and 2.97×10^{-3}. These standard deviations align with the relative 3% noise floor applied to the typical fractional normalized radiance levels of the scene. The normalized polarization residuals (Q/I) show minimal mean biases ranging from 3.62×10^{-5} to 1.22×10^{-3}, with standard deviations remaining stable across wavelengths, hovering around 0.003 (from 2.71×10^{-3} to 3.30×10^{-3}). This specification falls within the absolute noise floor of 0.005 assigned to the underlying Stokes parameters for the AOS polarimeter. As the simulations are performed in the principal plane, the U component is excluded from the analysis,

as it does not carry independent information in this geometry. The consistency of the method is confirmed by the absence of systematic biases across the eight spectral bands." (Page 30, line 639-648)

Comment 9: Line 665: These claims on AOD per species necessitate the reader to look at the supplement figure. I suggest adding it in the main text or limit this text to general statements.

Response: Clarified. We thank the reviewer for this suggestion. To keep the manuscript concise and focused on the primary results, we have opted to limit this discussion to general statements regarding the species-resolved AOD distribution rather than moving the supplementary figure into the main text.

The text around Line 665 has been revised as follows to ensure the main manuscript remains self-contained: "*AEROCHEMPro reproduces the expected spatial distribution of AOD at 532 nm across major aerosol species. Combustion-related species dominate over anthropogenic source regions, while mineral dust components are enhanced in arid regions such as Central Asia, and marine aerosols are prevalent over oceanic areas. Detailed species-resolved distributions are provided in Fig. S1 of the Supplement."* (Page 34, line 698-701)

Comment 10: Figure 9: LR and SSA are vertically varying parameters, but only one value is plotted here. What is the definition of reported LR and SSA here (and in section 3.2)? Please explain in the manuscript

Response: Clarification done. We thank the reviewer for identifying this ambiguity. The values plotted for SSA and LR in Figure 9 (and discussed in Section 3.2) represent the bulk, column-integrated optical properties derived from the column-integrated scattering, extinction, and backscatter coefficients at 532 nm.

To make this clear to the reader, we have updated the manuscript to explicitly use the term "total-column" or "column-integrated" in Section 3.2.1, the text describing Figure 9, and the captions for Figures 9 and 11. The specific text modifications are detailed below:

- Section 3.2.1 Title & Intro: Changed to "*Aerosol total-column optical properties*" and explicitly noted "*total-column single scattering albedo (SSA)*" and "*total-column lidar ratio (LR)*".
- Main Text (Figure 9 Description): Added the definition: "*Here, 'total-column' SSA and LR refer to bulk optical properties derived from column-integrated scattering, extinction, and backscatter coefficients at 532 nm.*" (Page 35, line 715-716)
- Figure Captions: Updated the captions for Figure 9 and Figure 11 to explicitly specify "*column-integrated*" or "*total-column*" parameters.

Comment 11: 711: Please change "surface-2km" to “between surface and 2km” or something similar

Response: Done. Thank you for the correction. We have revised the phrasing to improve readability. The text now reads: "*However, overestimations occur between the surface and 2 km, along with a simultaneous underestimation of water content linked to fine particles near coastal locations.*" (Page 36, line 744-745)