Response to Reviewer

We sincerely thank the reviewer for the evaluation of our work and the helpful suggestions to improve the clarity and rigor of the manuscript. Below, we provide detailed point-by-point responses. Comments from reviewers are shown in blue, **our responses in black**, and the corresponding changes made in the manuscript are highlighted in orange.

Comment 1:

The paper should clarify whether Single Scattering Albedo (SSA), asymmetry parameter (ASY), and relative humidity (RH) are necessary for the retrieval process. If these parameters are required, please briefly discuss how they can be obtained (e.g., from ancillary datasets, reanalysis products, or simultaneous measurements).

Response 1: **Auxiliary Parameters (SSA, AF, Reff, RH)**

Indeed, in our retrieval framework the parameters single-scattering albedo (SSA), asymmetry factor (AF), effective radius (Reff), and relative humidity (RH) are used as auxiliary (or "known") inputs to the machine-learning forward model. These parameters help constrain the spectral aerosol optical depth (AOD) mapping and thereby improve the stability and accuracy of the subsequent composition inversion.

In practice:

- For ground-based observations (e.g., from the AERONET network), SSA and other optical/size retrievals (including AF, Reff) are available as level-2 inversion products. The AERONET Version 3 algorithm provides derived SSA, phase-function parameters and asymmetry factor as part of its sky–scan measurements.
- For satellite applications, obtaining simultaneous SSA, AF and Reff directly from one instrument is more challenging. While satellite-based retrievals of SSA, AF, and Reff are becoming increasingly available, particularly from instruments such as POLDER, upcoming 3MI, and the PACE mission, these parameters typically require the synergy of multiple sensors or advanced inversion schemes. In contrast to ground-based systems like AERONET that provide consistent auxiliary data from a single instrument, satellite retrievals often involve merging heterogeneous sources, introducing additional uncertainty. Coordinated efforts across platforms and algorithm harmonization are thus critical for applying our framework to satellite observations on a global scale.

In our revised manuscript we have added:

L102-105: In addition to these direct sun measurements, AERONET provides inversion products that include single scattering albedo (SSA), asymmetry factor (AF), and effective radius (Reff), retrieved using sky radiance observations (Dubovik and King, 2000; Giles et al., 2019a). These parameters are useful for aerosol type discrimination and are used in this study.

Discusstion part:

L434- 446: Besides, our analysis highlights the importance of high-quality auxiliary optical parameters, particularly single scattering albedo (SSA), asymmetry factor (AF), and effective radius (Reff), in constraining aerosol composition retrievals. In ground-based settings, instruments such as AERONET offer reliable inversions of these parameters, enabling accurate component estimation when combined with spectrally resolved AOD. While satellite retrieval of these quantities remains more challenging. Current and upcoming satellite sensors can provide some of this information, but typically require multi-angle, multi-spectral, or polarimetric measurements, along with advanced inversion algorithms. For example, the POLDER instrument aboard PARASOL enabled global retrievals of SSA, Reff, and AF through polarization-based algorithms such as GRASP and RemoTAP (Dubovik et al., 2011b; Hasekamp and Landgraf, 2007). Upcoming missions like 3MI (on MetOp-SG) and NASA's PACE will carry advanced polarimetric imagers and hyperspectral sensors to further improve retrievals of aerosol microphysical and optical properties (Werdell et al., 2019; Gao et al., 2021a). However, these satellite-based parameters often originate from different sensors and require cross-platform coordination, unlike AERONET which provides all relevant quantities from a single instrument. Therefore, applying the proposed retrieval framework to satellite observations may involve higher uncertainty and greater dependence on auxiliary data. Future improvements in satellite instrumentation and algorithm synergy will help extend this framework from ground-based to global applications.

Comment 2:
The manuscript claims that AOD in infrared (IR) wavelengths provides additional information on aerosol composition. Please elaborate on this point—for example, by explaining how IR absorption features are linked to specific aerosol types (e.g., dust, organic carbon) or how they complement visible/UV observations.

Response 2:
We appreciate the reviewer's request to clarify the role of infrared AOD in aerosol composition retrieval. As illustrated in the Figure 2, our simulation using MOPSMAP shows that different aerosol types exhibit distinct spectral signatures, especially in the shortwave infrared (SWIR) region.

While many aerosol types (e.g., black carbon, dust, insoluble organics) are spectrally similar in the visible range (440–870 nm), their normalized AOD spectra begin to diverge significantly beyond 1.5 μm. This behavior is driven by differences in size distribution and refractive index: for instance, sea salt retains a high AOD in the IR due to its coarse-mode scattering efficiency, whereas sulfate drops off more steeply. Similarly, dust exhibits enhanced scattering at longer wavelengths compared to black carbon, which remains relatively absorbing across the spectrum.

L305-309: These differences suggest that including SWIR wavelengths helps distinguish composition types that would otherwise be difficult to separate using visible AOD alone. The use of IR channels, therefore, increases the information content available to the retrieval, especially for distinguishing aerosols with similar visible properties but different infrared behaviors. We have clarified and expanded this point in the revised manuscript to better reflect the physical motivation for including SWIR wavelengths.

Comment 3:
The text refers to MOSMAP as a “radiative transfer model,” but it appears to be a bulk aerosol optical property calculator based on size distribution and refractive index inputs. Please correct this terminology. Additionally, the study relies solely on Mie scattering, neglecting non-spherical scattering methods (e.g., T-matrix for dust). Since dust aerosols are often nonspherical, this simplification may introduce errors. A brief discussion on this limitation and its potential impact should be included.

Response:
We agree with the reviewer’s correction and have corrected the terminology and now refer to MOPSMAP as an aerosol optical property calculator.

Regarding particle shape, we acknowledge that assuming spherical aerosols may lead to biases, especially for mineral dust. Determining particle shape is a complex task that typically requires active remote sensing techniques such as lidar to provide depolarization or polarization ratios. In our previous work (Ji et al., 2023), we demonstrated that joint FTIR and lidar observations can help constrain aerosol properties more comprehensively. Incorporating shape information into future retrieval frameworks, particularly using lidar-derived parameters, is a promising direction we plan to explore.

We have added a paragraph in the Discussion section to acknowledge this limitation and its implications:
L427- 432: In this study, the use of Mie theory assumes spherical aerosol particles, which may introduce biases, particularly for non-spherical particles such as mineral dust. Determining aerosol shape is a complex problem that cannot be addressed solely through passive radiometry. In practice, characterizing particle asphericity requires additional measurements, such as polarization or depolarization ratios from lidar or radar. Our previous work (Ji et al., 2023) has demonstrated the feasibility of combining FTIR and radar observations to jointly constrain aerosol properties. Therefore, incorporating aerosol shape information through active remote sensing is a promising avenue for future improvement of the retrieval framework.

Comment 4:
The Optimal Estimation Method (OEM) requires prior information and its associated covariance matrix. The manuscript should clarify:
(i) Whether prior estimates are sourced from MERRA-2 or other datasets.
(ii) How the covariance matrix of the prior is defined (e.g., based on climatological variability, instrument uncertainty, or empirical assumptions).

Response:
Thank you for highlighting this. We have added clarification in Section 3.4.

L245- 254: The a prior vector x_a is derived from the MERRA-2 monthly mean aerosol component fractions at the same time and location as the FTIR observations. The a prior covariance matrix S_a is set as a diagonal matrix with variance 0.01 (i.e., standard deviation of 0.1) for each aerosol component. This reflects a relatively loose prior constraint, allowing the retrieval to be primarily informed by the spectral AOD observations while maintaining physical plausibility. For the measurement error covariance matrix S_γ, we distinguish between visible and shortwave infrared (SWIR) wavelengths. For visible bands (AERONET-like observations), we adopt 0.01 as the standard deviation, consistent with the reported uncertainty of AOD retrievals from AERONET. For the infrared bands (SWIR), we adopt 0.02 as the standard deviation, based on reported uncertainties from Barreto et al. (2020) and Alvárez et al. (2023), who applied Langley calibration for FTIR-based AOD measurements in the SWIR region. All uncertainties are assumed to be spectrally uncorrelated, and S_γ is constructed as a diagonal matrix. This assumption has been clarified in the revised manuscript.