# Replies to Referee #2

We would like to sincerely thank the Referee for their careful reading of our manuscript and for their constructive and helpful comments. Their suggestions have helped us to significantly improve the clarity, structure, and scientific depth of the paper. In the revised version, we have addressed all comments point by point. Changes in the text are clearly marked in the manuscript *with red fonts*.
Below, we provide detailed responses (*brown fonts*) to each reviewer's comment (*black fonts*) and indicate where the corresponding revisions have been implemented.

## General comments:

The manuscript documents the exceptional aerosol conditions over Athens during the summer 2023 Greek wildfire season, focusing on two major episodes (18–21 July and 22–25 August). It combines MODIS fire detections, FLEXPART retroplume simulations, multi-wavelength Raman/polarization lidar, AERONET sun–sky–lunar photometry, in-situ PM/BC and ACSM chemistry, and Brewer UV spectral measurements to characterize aerosol vertical structure, optical/microphysical properties, and the associated impact on surface UV at 320 nm (treated as a proxy for UV-B). Reported conditions include AOD up to ~1.2, high fine-mode fractions, SSA spanning ~0.85–0.98, PLDR variability consistent with smoke–dust mixing, and surface 320-nm irradiance reductions up to ~50% during peak smoke.

My recommendation is major revision. The dataset and the synergistic concept are strong, but several central methodological choices (especially the UV/SSA retrieval setup, cloud screening, and uncertainty propagation across the chain) are not yet documented or stress-tested enough to support some of the stronger interpretations.

## Specific comments:

<u>Comment:</u>

The paper equates 320-nm irradiance with "UV-B impact" but does not demonstrate how well 320 nm tracks band-integrated UV-B (290–315 nm) variability under changing ozone and aerosol conditions.

<u>Reply:</u>

At lines 166 – 170 in the original version of the manuscript we clarify that:

"The 315 – 325 nm wavelength range has been chosen because at these wavelengths, for relatively high aerosol loads (e.g., during wildfire or dust events), the effect of aerosols is dominant over the effect of ozone. For shorter wavelengths (<315 nm) variations and uncertainties in total ozone induce higher uncertainties regarding the quantification of the impact of aerosols. We consider that the impact of aerosols at these wavelengths is representative for their impact on UV-B (290 – 315 nm integral)."

The following lines have been added in the manuscript.

"The impact of aerosols on the 290 – 315 nm integral could be masked, at least partially (see e.g., Fountoulakis et al., 2016;2019), by the effect of ozone, and this is why we choose to show the results for the 320 nm irradiance. Given that most of the attenuation by ozone takes place in the stratosphere while interaction of the UV-B radiation with aerosols happens at the lowest 2 -3 km in the troposphere, complex interactions between UV-B, aerosols and ozone under heavy aerosol load are not expected to impact the variability in UV-B, at least significantly.

Differences between the variability (caused by aerosols) in the 320 nm irradiance and the 290 – 315 nm integral are expected to be mostly due to the corresponding differences in aerosol optical properties (AOD, SSA), which would introduce a slightly larger variability for shorter wavelengths. The differences, however, are estimated to be small, within the uncertainty limits in the UV measurements."

Comment:

The UV closure relies on cloud-free simulations, yet the lidar time–height plots clearly show cloud formation during key windows; without explicit cloud/thin-cloud screening rules and the retained time stamps, the UV comparisons are hard to trust.

Reply:

The reviewer is correct. In the revised version of the manuscript, all measurements were visually inspected in conjunction with lidar time–height plots, and data affected by the presence of clouds or thin clouds were excluded. The discussion and panels (e)–(g) of Figure 12 already refer to cloud-free cases, while measurements acquired under cloudy conditions have now been removed from panels (a)–(d) for clarity. The updated Figure 13 reflects these changes.

We note, however, that a small influence from thin or subvisible cirrus clouds cannot be entirely excluded in certain cases, as our lidar system does not perform scanning observations and may not always detect high-level cirrus layers if they are optically very weak or above the instrument's detection range.

We have added the following lines in Section 3.7: "All analyzed periods correspond to cloud-free conditions, as confirmed by visual inspection of the lidar spatio–temporal profiles and Brewer data. Measurements affected by cloud or thin cloud presence were excluded to ensure reliable comparison with the clear-sky radiative transfer simulations. However, we note that a small influence from optically thin or subvisible cirrus clouds cannot be entirely ruled out in certain cases, since our lidar system operates in a fixed vertical configuration and may not detect very weak or high-level cirrus layers."

Comment:

The SSA(320) retrieval uncertainty is treated mainly via ±5% irradiance bounds, but AOD(320), surface albedo, aerosol vertical distribution, and phase function (dust vs smoke) can easily dominate the error budget; some structured sensitivity tests (or an error propagation table) are needed.

Reply:

The reviewer is again correct. We thank him/her for this comment. As discussed in Fountoulakis et al. (2019), the impact of uncertainties in AOD, surface albedo, etc. is more significant than the impact of uncertainties in the irradiance measurements. Nevertheless, for relatively high AOD (340 nm AOD > 0.3) the effect of the latter uncertainties becomes less significant, and comparable to the effect of uncertainties in the irradiance. The phrase:

"Even assuming irradiance values that are 5% above the measured irradiances, results in this case in SSA values are below 0.9."

has been replaced with the following one:

"The boundaries shown in Figure 13 do not represent the full range of uncertainties in the retrieval of the SSA. Additional uncertainty in the order of 0.1 must be considered due to uncertainties in the inputs that were used for the LibRadtran simulations (AOD, surface albedo, aerosol profile, etc.) (e.g., Fountoulakis et al., 2019). Thus, the results presented here should be treated with caution. Nevertheless, they provide a strong indication for the validity of the findings of previous studies showing that dust particles can be more absorbing in the UV-B compared to smoke particles."

Comment:

AOD extrapolation to 320 nm using Ångström behavior (e.g., 340–440 nm) can be biased in coarse-mode/dust or mixed cases (spectral curvature); the "~0.05 agreement" should be broken down by event type (smoke / dust / mixed), not just pooled statistics.

Reply:

We have added the following in the manuscript (please refer to Section 2.4):

For the case studies, the differences between the 320 nm AOD retrieved from the Brewer and the extrapolated AOD (from 340 nm to 320 nm using the Ångström exponent) derived from CIMEL are generally smaller for the dust case (0.02–0.05) compared to the smoke case (0.05–0.08). Nevertheless, a more comprehensive analysis is required to determine whether these differences reflect limitations of the Ångström approximation in capturing the spectral curvature of AOD at short wavelengths or arise from the overall uncertainty of the AOD retrievals.

Comment:

There is an internal tension that needs clarification: the manuscript states the least UV attenuation occurs on the dust day (23 July), while also retrieving very low SSA(320) ~0.75–0.8 for dust; it is not clear whether the comparison is for global vs direct vs diffuse irradiance and whether residual cloud effects or geometry explain this.

Reply:

The statement here was unclear. As the reviewer can notice in Figure 2 the AOD on July 23$^{rd}$ is the lowest among the three days. We thank the reviewer for noticing that. The paragraph (lines 616 - 624) has been replaced by the following one:

"These patterns are also supported by the diurnal plots in Figure 13 (e-g), which present UVB irradiance measurements at 320 nm for three selected days, each representing a different aerosol event discussed in Section 3: 21 July (smoke + dust), 23 July (dust-dominated), and 24 August (smoke-dominated). On 21 July, elevated AOD resulted from the combined presence of smoke and dust, while 23 July was characterized by high AOD due to the presence of dust. The most extreme UVB attenuation occurred on 24 August, coinciding with the highest AOD values of the period, driven by smoke. In contrast, the least attenuation was observed on July 23rd, corresponding to the lowest AOD among the three days. These differences align with aerosol optical characteristics shown in Figure 3: 21 July featured high Ångström exponent (AE > 1) and fine-mode fraction (FMF ≈ 0.6), indicating mixed aerosols, 23 July had low AE (~0.5) and FMF (~0.3), consistent with coarse-mode dust dominance, while 24 August exhibited strong fine-mode influence, with AE around 2.0 and AOD at 340 nm exceeding 1.0."

Comment:

The microphysical inversion assumes a wavelength-independent complex refractive index across 355–1064 nm; this is a strong assumption for smoke (possible brown carbon) and dust, and its effect on retrieved $m_i$, $R_{eff}$, etc. should be quantified.

Reply:

Yes, the assumption of spectrally independent CRI may affect the result. Number of input data from 3+2 data set is insufficient for retrieval the spectrally dependent CRI. We can only estimate the uncertainty. Analysis of influence of spectrally dependent imaginary part of dust was reported in (Atmos. Chem. Phys., 16, 7013–7028, 2016 www.atmos-chem-phys.net/16/7013/2016/ doi:10.5194/acp-16-7013-2016)

For the dust near the source of origin (West Africa) introduction of model spectral dependence, at low altitudes, could introduce up to 50% change in values of volume and effective radius. But spectral dependence of imaginary part in that episodes was noticeable because the backscattering Angstrom exponent (BAE) 355-532 was less than -0.5. In long transported dust this effect is less because BAE is about zero. The estimated corresponding uncertainty is less than 20%.

For smoke, this effect is much less (because particles are small). This statement is corroborated by comparison of extinction-to-volume conversion factors for smoke obtained from AERONET and retrieved from 3+2 lidar inversion (Veselovskii, I., Hu, Q., Goloub, P., Podvin, T., Dubois, G., Kolgotin, A., and Korenskii, M.: Impact of water uptake on fluorescence of atmospheric aerosols: insights from Mie–Raman–fluorescence lidar measurements, Atmos. Meas. Tech., 18, 6039–6051, https://doi.org/10.5194/amt-18-6039-2025, 2025), which just coincide.

Comment:

Treating particles as spheres for PLDR<10% and spheroids otherwise is too binary for mixed layers; at minimum, show threshold sensitivity (e.g., 5/10/15%) or discuss a continuous mixing approach.

Reply:

Effect of particle shape on retrieval of dust and smoke properties was analyzed in publication (Veselovskii, I., P. Goloub, T. Podvin, D. Tanre, A. da Silva, P. Colarco, P. Castellanos, M. Korenskiy, Q. Hu, D.N. Whiteman, D. Pérez-Ramírez, P. Augustin, M. Fourmentin, A. Kolgotin: Characterization of smoke/dust episode over West Africa: comparison of MERRA-2 modeling with multiwavelength Mie-Raman lidar observations, Atm. Meas. Tech. 11, 949–969, 2018.)

Inversion of the same data with spheres and spheroids leads to similar volume densities. For both smoke and dust particles difference is less than 30%. Similar difference is for effective radius. When dust is mixed with pollution this difference will be less. We estimate uncertainty of volume and effective radius retrieval to be less than 20%, when particles with PLDR<10% are treated as spheres and as spheroids otherwise.

Comment:

POLIPHON mass profiles depend on fixed parameters (e.g., smoke density 1.35 g cm-$^{3}$, variability allowances, “non-collocated still effective”); this may be reasonable for stable dust, but wildfire plumes are temporally/vertically variable—please justify applicability for the smoke cases and propagate these choices into uncertainty bars.

Reply:

The method remains effective even when lidar and photometer data are not strictly collocated, especially for stable dust events. Total uncertainty in retrieved mass concentrations is ~36–40 % and assumed mass densities (±20 %). Volume-to-AOD ratios may vary by up to 10 % for dust and 20 % for smoke (Ansmann et al., 2012; Ansmann et al. 2019).

In our case, the lidar and sun–sky photometer are collocated on the same building, ensuring consistent column and profile measurements and minimizing spatial mismatch. Therefore, the reported total uncertainty (~ 36–40 %) primarily reflects the combined variability in assumed aerosol properties and retrieval parameters rather than site separation effects.

Furthermore, we also added in Section 2.6.4 the lines: “In our case, the lidar and sun–sky photometer are collocated on the same building, ensuring consistent column and profile measurements and minimizing spatial mismatch.

Therefore, the reported total uncertainty (~ 36–40 %) primarily reflects the combined variability in assumed aerosol properties and retrieval parameters rather than site separation effects."

Comment:

FLEXPART configuration needs better alignment with observations: if aerosol layers reach ~5–6 km but the release layer is limited to 0–4 km, source attribution may be incomplete; sensitivity to release height and back-trajectory duration should be demonstrated.

Reply:

We thank the reviewer for this useful comment. The FLEXPART backward simulations were configured with release heights between 0–4 km to represent the altitude range of the aerosol layers observed by lidar in both case studies. Most of the retrieved layers were located within these limits. For the few instances where aerosol backscatter extended up to ~5 km, additional sensitivity tests using an extended-release layer (0–5 km) showed no substantial change in the spatial source patterns or relative source contributions. Therefore, the 0–4 km configuration provides a representative and robust alignment with the observed aerosol layers while maintaining computational efficiency. This clarification has been added to Section 2.6.3.

We also added the following lines in Section 2.6.3: "In our case, most lidar-retrieved aerosol layers were located below 4 km, well represented by the 0–4000 m release range. For a few events where layers extended up to 5 km, sensitivity tests using an extended 4000–5000 m release showed negligible differences in the source–receptor patterns, confirming that the chosen configuration adequately captures the observed aerosol transport."

Comment:

The smoke/dust/mixed attribution is mostly qualitative across multiple indicators (AE, FMF, PLDR, LR); a single reproducible classification rule (even simple thresholds with rationale) would make the "synergy" claim much stronger.

Reply:

We appreciate the reviewer's valuable observation. In the revised version, we now explicitly define a quantitative and reproducible classification rule for aerosol type attribution, consistently applied across all indicators.

For column-integrated optical properties, we use threshold values of AE > 1.2 and FMF > 0.8 for Smoke (fine-mode dominated), AE < 0.8 and FMF < 0.5 for Dust (coarse-mode dominated), with all other cases labeled Mixed. These align with AERONET climatologies for fine/coarse-mode separation.

For lidar profiles, we apply established thresholds from Ansmann et al. (2011, 2019), Mamouri & Ansmann (2014), and Papagianopoulos et al. (2018):

- Smoke: $LR_{355}$, $LR_{532}$ $\lesssim$ 50-70 sr (low absorption) and $\delta_{532} \lesssim$ 0.05-0.10 (non-depolarizing)
- Dust: $LR_{355}$, $LR_{532}$ $\gtrsim$ 40-60 sr (typical dust) and $\delta_{532} \gtrsim$ 0.20-0.30 (strongly depolarizing)
- Mixed: intermediate values

We have also included a new Supplementary Figure S8 showing the resulting classification alongside the AERONET and lidar data. This quantitative, literature-supported framework ensures objectivity and reproducibility, strengthening the synergy between the photometric and lidar observations.

Moreover, we also added the following lines in Section 3.6: "A quantitative classification of the aerosol types based on established lidar ratio and depolarization thresholds (Figure S8) further supports this interpretation, distinguishing dust-dominated, smoke-dominated, and mixed aerosol layers during this period."

Comment:

Lidar-retrieved and AERONET-retrieved effective radius differ substantially; the discussion currently stops at "different sensitivities," but the paper should compare layer-integrated vs column quantities and explain whether the discrepancy affects downstream radiative interpretation.

Reply:

We thank the reviewer for this comment. We have expanded the discussion and quantified the comparison between lidar- and AERONET-retrieved effective radius. Specifically, we calculated correlations and bias statistics for the overlapping dataset:

- Pearson correlation: R = 0.39 (p = 0.033)
- Spearman correlation: R = 0.54 (p = 0.002)
- Root-mean-square error (RMSE): 0.213 µm
- Mean bias (Lidar – Sun-photometer): −0.132 µm

The moderate correlations and small negative bias indicate that lidar and AERONET retrievals capture consistent temporal variability, but lidar tends to slightly underestimate the column-integrated effective radius. This discrepancy is expected because lidar retrievals provide layer-resolved effective radius within aerosol layers and AERONET effective radius is a column-integrated quantity, weighted by optical depth.

To illustrate this, we included a scatter plot of lidar vs. AERONET effective radius in Supplementary Figure S5. These results support that the difference primarily reflects the different retrieval domains (layer vs column) rather than a systematic disagreement, and that the discrepancy is small enough to have limited impact on downstream radiative interpretation in our study.

We also added the following lines in Section 3.6: "To quantify the consistency between lidar and sun photometer retrievals, we calculated correlation and bias statistics (Figure S5). The Pearson correlation coefficient is 0.39 (p = 0.033) and the Spearman correlation is 0.54 (p = 0.002). The root-mean-square error (RMSE) is 0.213 µm, with a mean bias (Lidar − Sunphotometer) of −0.132 µm. These results indicate that the two retrievals capture consistent temporal variability, although lidar slightly underestimates the column-integrated $R_{eff}$. The RMSE and mean bias suggest that column-integrated radiative calculations using either product would differ by ~0.2 µm, which is within the expected uncertainty of aerosol microphysical retrievals."

Comment:

Statements about "strong absorption" lean heavily on retrieved $m_i$ and UV-SSA; please tighten the cross-validation with independent constraints (AERONET SSA variability, eBC source apportionment, and ACSM organics) so the absorption narrative is not driven by one retrieval stream.

Reply:

In response to the comment, we have added the following statement in Section 3.6 to strengthen the cross-validation of aerosol absorption:

“These absorption features are supported by independent observations: AERONET SSA at 440 nm decreased to ~0.85 during peak wildfire events, eBC concentrations measured at the surface peaked concurrently with elevated smoke layers, and ACSM measurements indicate a high fraction of organics. Together, these constraints confirm that the strong absorption is real and not driven solely by the lidar retrievals.”

Comment:

The manuscript links elevated free-tropospheric smoke layers (2–5 km) with surface PM/chemistry peaks, but the coupling mechanism is not demonstrated; a short analysis using BLH, timing consistency, and evidence for downward mixing would help separate transported impacts from local/BL contributions.

Reply:

In response to the comment, we have expanded the discussion to clarify the coupling between elevated smoke layers and surface aerosol enhancements. Although the lidar observations were conducted at NTUA and the in-situ measurements (PM and chemical composition) at Demokritos, the two sites being approximately 10 km apart are considered regionally representative of the Athens basin. ERA5-derived planetary boundary layer height (PBLH) data were analyzed to assess vertical coupling. Supplementary Figures S3–S4 show that, during several episodes, the PBLH increased sufficiently to intersect the lower portion of the 0.5–4 km smoke layers detected by lidar. These periods coincided with surface $PM_{2.5}$ and eBC peaks, indicating partial downward mixing from the free troposphere to the surface. This additional analysis, described in Section 3.2 and illustrated in the Supplement, supports that part of the observed surface enhancements can be attributed to transported smoke, while acknowledging concurrent contributions from local boundary-layer sources.

We also included the following lines in Section 3.2:

“To further examine the coupling between elevated smoke layers and surface aerosol enhancements, ERA5-derived planetary boundary layer height (PBLH) data were analyzed together with simultaneous lidar and in-situ observations (Supplementary Figures S3–S4). During July and August episodes, the PBLH increased during daytime, occasionally intersecting the lower portion of the elevated aerosol layers, coinciding with surface $PM_{2.5}$ and eBC peaks. These findings indicate that part of the surface aerosol enhancement can be attributed to downward mixing of transported smoke, in addition to local boundary-layer contributions.”