Responses to Referee #1

Thank you for reading the manuscript and providing very useful comments and suggestions to improve the paper. The referee comments are in black, while our replies are in blue. The modifications in the revised manuscript can be found in the track change version of manuscript.

Just two remarks:

I still found "Flament" on three occasions. I think "Flamant" is correct. Please, scan the text.

AR: Thanks for the reminding. We checked the "Flament" and "Flamant" throughout the manuscript and confirmed that there are two different "Flament" and "Flamant": Flamant et al. (2008) and Flament et al. (2022) indicate Pierre H. Flamant, who authored the paper "ADM-Aeolus retrieval algorithms for aerosol and cloud products" and the report "Aeolus L2A Algorithm Theoretical Baseline Document, Particle optical properties product". Flament et al. (2021) indicate Thomas Flament, who authored the paper "Aeolus L2A aerosol optical properties product: standard correct algorithm and Mie correct algorithm".

References:

Flamant, P. H., Cuesta, J., Denneulin, M.-L., Dabas, A., and Huber, D.: ADM-Aeolus retrieval algorithms for aerosol and cloud products, Tellus A, 60, 273–286, https://doi.org/10.1111/j.1600-0870.2007.00287.x, 2008.

Flamant, P., Dabas, A., Martinet, P., Lever, V., Flament, T., Trapon, D., Olivier, M., Cuesta, J., and Huber, D.: Aeolus L2A Algorithm Theoretical Baseline Document, Particle optical properties product, Tech. rep., ESA, version 6.0, https://earth.esa.int/eogateway/documents/20142/37627/Aeolus-L2A-Algorithm-Theoretical-Baseline-Document.pdf (last access: 28 May 2026), 2022.

Flament, T., Trapon, D., Lacour, A., Dabas, A., Ehlers, F., and Huber, D.: Aeolus L2A aerosol optical properties product: standard correct algorithm and Mie correct algorithm, Atmos. Meas. Tech., 14, 7851–7871, https://doi.org/10.5194/amt-14-7851-2021, 2021.

In line 401 of the version with tracked changes I see "transported from the US": to where?

You could also write "advected from the US".

AR: Thanks, revised as "This smoke layer observed over Europe was found to have been advected from the US".

Responses to Referee #2

Thank you for reading the manuscript and providing very useful comments and suggestions to improve the paper. The referee comments are in black, while our replies are in blue. The modifications in the revised manuscript can be found in the track change version of manuscript.

We have carefully considered all the comments and technical corrections, and the manuscript has been revised accordingly. Our responses to the remaining concerns are provided below.

1. Figure 3: I'm afraid I don't understand parts of this flowchart

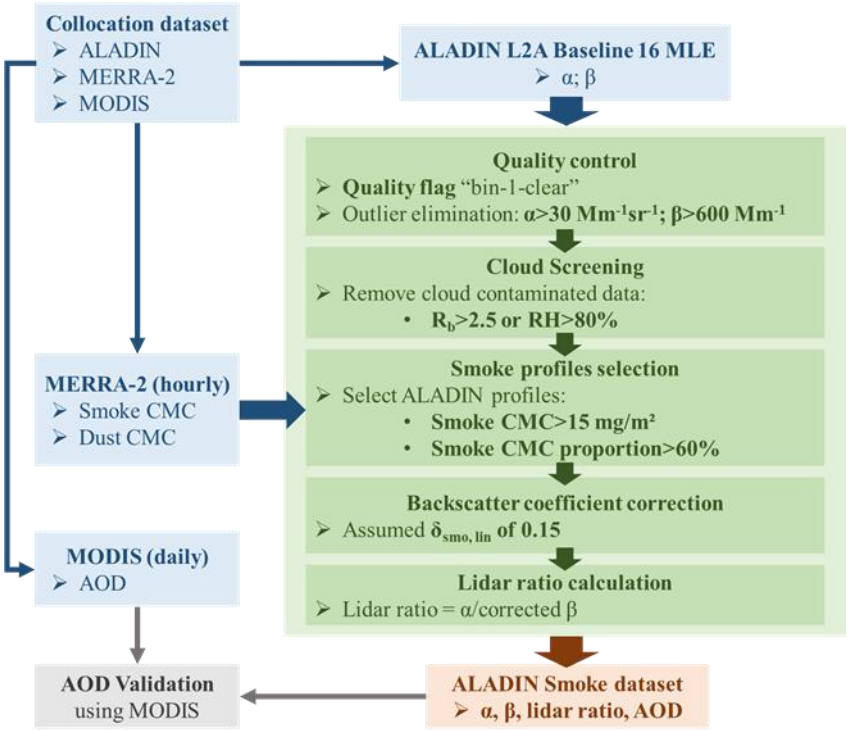
(1) I'm confused by the apparent feedback loop between the ALADIN L2A data and the MERRA-2 + MODIS data.

(2) why are both MERRA-2 and MODIS apparently inputs to the smoke section process? according to line 248, MODIS is used for validation, not derivation (e.g., identifying qualifying profiles). so perhaps the MERRA-2 and MODIS contributions should be in totally separate boxes and not embedded in a single pale blue box?

(3) can you provide more detail in the "Validation" box? e.g., "Validation using MODIS"

(4) based on my (perhaps wildly incorrect) understanding of the text, the flowchart should look like Figure3Revisions.png attached to the previous comment

AR: Thanks for pointing this out. We have restructured and revised the flowchart according to your suggestions to improve its clarity and avoid potential misunderstanding. The revised Fig.3 is shown below. It begins with the "Collocation dataset", which then branches into three datasets, each leading to the corresponding processing procedures. The overall layout follows a left-to-right and top-to-bottom structure, consistent with common reading conventions, making the workflow easier to follow.



2. Line 264, "Cloud screening": please state how frequently smoke layers were detected above clouds in this dataset.

AR: Thanks for the suggestion. Combining with Comment 6, we have provided additional evidence, beyond the "smoke layers above clouds" shown in Fig. 7, to support the statement that "the smoke layers appeared above clouds when transported to Europe". Specifically, we examined the complete ALADIN dataset and added the following description: "On 20 and 21 September, the ALADIN observations indicated that the smoke layers were located above the cloud layers. Across the complete dataset, five of the six smoke layers identified in the ALADIN cross-sections over Europe during these two days were found above clouds. This suggests that the smoke plume was predominantly above clouds upon reaching Europe." This description has been added to the last part of Section 5.1.1.

3. Line 321, "are unlikely to originate from typical aerosol cases": true. but I don't believe that these intense wildfires represent a "typical aerosol case".

AR: Thanks. The sentence has been revised as "The principle for threshold setting is that values exceeding the thresholds are unlikely to fall within the typical range of aerosol optical properties".

4. Line 630: "In order to derive a smoke dataset of extinction coefficient, backscatter coefficient and lidar ratio from Aeolus, a collocation dataset comprising Aeolus products, MODIS AOD, and MERRA-2 smoke column mass concentration (CMC) was established." CALIOP depolarization ratios were essential for retrieving total backscatter coefficients, extinction coefficients, and lidar ratio. but for some reason, CALIOP is not included in this list.

AR: Thanks for pointing this out. We added a sentence "The backscatter coefficient correction was achieved with the aid of CALIOP depolarization ratios." after that description.

5. Line 645: repeating a comment from my initial review, "It's still not clear to me how ALADIN alone can distinguish between thin cloud and dense aerosol". nor is it obvious to me which "multi-source data" (other than perhaps CALIOP) would provide dispositive information.

AR: We acknowledge that ALADIN alone cannot distinguish between thin clouds and dense aerosol layers. However, here we aim to highlight the capability of the "Aeolus smoke dataset" developed in this manuscript, which was derived with the assistance of MERRA-2 CMC and CALIOP depolarization ratios. Based on this dataset, smoke aerosol layers were identified above clouds, providing stronger evidence than MODIS observations alone. Therefore, we have retained this statement in the manuscript.

6. Line 654: again, while I do not doubt the truth of this claim, the authors are making the assertion without providing direct evidence (e.g., see my request on line 264).

AR: Thanks. We have added additional evidence, as described in our response to Comment 2.

Responses to Referee #3

Thank you for reading the manuscript and providing very useful comments and suggestions to improve the paper. The referee comments are in black, while our replies are in blue. The modifications in the revised manuscript can be found in the track change version of manuscript.

I thank the authors for a conscientious revision. They have addressed the comments from all three referees and made substantial changes to the manuscript. The title has been narrowed. The new dedicated uncertainty section and a methodology flowchart address my comments. The results section was restructured. The framing of the transport interpretation is more cautious and descriptive rather than dynamical. As most of my concerns have been addressed satisfactorily, my recommendation is minor revision. The manuscript is close to acceptable, with a small number of technical corrections still needed.

My major comments on the transport interpretation have been addressed by adjusting the scope rather than by adding dynamical analysis. The title of the paper focusses now on aerosol characterization. Some speculative claims have been softened. The synoptic and isentropic interpretation with Aeolus winds can be followed up in a future paper. I find this suitable for a characterization study, even if it is a reframing rather than a strengthening of the transport interpretation. A similar consideration applies to the ACP versus AMT question: the scope focused on characterization of aerosol properties supports publication in ACP, but that decision is for the editor to make.

The transport interpretation and the representativeness of the measurements for different transport stages have been partially addressed. The word "confirmed" has now been replaced by more cautious language, and the cross-section claims are better qualified, which is welcome. Part of the interpretation still rests, however, on a limited trajectory calculation. Figure 7 shows a small set of nine forward and nine backward trajectories released from three locations at three heights at a single time, 10:00 UTC on 18 September 2020. A set of this size can be useful as illustrative support for the general transport pathway, but it is not statistically robust enough to establish the dynamics or representativeness of the plume. A Lagrangian particle dispersion approach with a larger ensemble would be required for a proper dynamical transport analysis. I am not asking for additional simulations, since the paper is now framed as a characterization of aerosol properties using combined remote sensing measurements. However, the trajectories should be presented consistently as illustrative support for the general transport pathway, not as evidence establishing the representativeness or dynamics of the plume. This applies throughout the manuscript, including the abstract, where "HYSPLIT simulations support the representativeness of these layers across the transport" still seems too strong. A formulation such as "HYSPLIT simulations illustrate the general transport pathway connecting these layers" would be more appropriate, and parallel statements in Sections 5.1.2 and 6 should be softened in the same way.

AR: Thanks for the suggestion. We have softened the statement in the abstract, Section 5.1.2 and 6 accordingly.

The plume separation concern is partly addressed by the new Section 5.2.2, which quantifies the two branches separately and is a clear improvement. The mechanisms remain inferred rather than demonstrated, but the wording is now more cautious. The requested uncertainty section (new 4.3) and the reproducibility guide (Figure 3 plus Sections 4.1 to 4.3) are good additions, and the switch from standard deviation to SEM in Table 2 is appropriate.

In Section 5.1.2, the sentence "However, they only provide a limited depiction of the entire smoke plume, which can extend over several thousand kilometers" is useful and should be retained because it refers to

the selected ALADIN cross sections. I suggest adding a second sentence after it: “The accompanying HYSPLIT trajectories should likewise be regarded as illustrative, since they are based on a limited number of releases from one time and from a small number of locations and altitudes.”

AR: Thanks, added.

My minor comments were all addressed: the novelty statement, the justification for choosing the MLE 90 km product, the Ångström exponent (now 0.5 from Hu et al. rather than the fixed 0.2, applied consistently), the cautious phrasing on smoke above cloud, the Figure 9 caption, and Table A1.

A few items remain before acceptance, listed roughly in order of importance.

First, Equations (3) and (4) are inconsistent. Both express the first order propagation of the linear depolarization uncertainty into, respectively, the corrected backscatter coefficient and the lidar ratio. Working from Equations (1) and (2), the corrected backscatter is $\beta_{smo} = \beta_{Aeolus}(1+\delta)/(1-\delta)$, so its derivative with respect to δ is $2\,\beta_{Aeolus}/(1-\delta)^2$, and the denominator of Equation (3) should therefore be squared. Equation (4) already carries the corresponding $(1+\delta)^2$ and is correct, which makes the missing square in Equation (3) an internal inconsistency. The numerical effect is minor and does not change the conclusions, but the equation should be corrected. I also suggest the authors state explicitly that σ_β and σ_L are first order propagated standard deviations, as this is not currently defined.

AR: Thanks. Sorry for the mistake in Equation (3). We have revised the equation and added the definition of σ_β and σ_L as the “first order propagated standard deviations”.

Second, the uncertainty section should acknowledge methodological uncertainty and not only measurement noise. Section 4.3 states that steps (1) to (3) do not introduce uncertainties beyond those inherent in the Aeolus observational data. This is too strong. The outlier thresholds, the cloud screening thresholds on Rb and RH, and the MERRA-2 based smoke profile selection are methodological choices that change which bins and profiles enter the statistics, and they can therefore affect the reported mean optical properties and trends, even if they leave the underlying measurement noise unchanged. A full sensitivity study is not necessary, but the section should recognize this source of uncertainty rather than set it aside.

AR: Thanks for the suggestion. We added the statement “Nevertheless, the bin selection itself is subject to the methodological uncertainty because it relies on the threshold values of Rb, RH and smoke CMC, all of which are associated with their own uncertainties. Consequently, these uncertainties may influence the retrieved mean optical properties.” in the first paragraph of Section 4.3, to avoid ignoring methodological uncertainty.

Third, several conclusions are phrased more strongly than the evidence supports. In particular, the statement that Aeolus can provide reliable observation of a smoke transport event is too categorical, since the event validation relies mainly on an AOD comparison after wavelength conversion, which does not directly validate the corrected backscatter or the lidar ratio. A more measured phrasing, such as "provides useful and broadly consistent observations," would be safer. The "first use" claim and the smoke above cloud statements would benefit from similar tightening. More generally, since the paper is framed as an aerosol characterization study, the transport conclusions should remain explicitly observational, and the link between the lidar ratio increase over Europe and higher relative humidity should continue to be presented as an interpretation supported by co-variation rather than as a demonstrated mechanism.

AR: Thanks. Softened and tightened the statements accordingly. All the revisions are shown as below:

"In conclusion, we demonstrated that Aeolus can ***provide useful and broadly consistent observations*** of smoke transport event with the synergy of multi-platform data."

"To our knowledge, this paper presents the first use of ***ALADIN*** for observations and analyses of a large-scale transatlantic tropospheric smoke transport."

"The smoke layers ascended from approximately 4.4 km above the US to over 6 km above the Europe and ***were observed*** above clouds upon reaching Europe."

"The initially decreasing and then increasing lidar ratios ***are likely associated with*** the aging process and the hygroscopic growth of smoke particles, individually."

"As revealed by the cross-sections, the declining trend of the lidar ratio from 62 sr on 14 September to 44 sr on 19 September ***may be attributed to*** the aging process of smoke aerosols. Subsequently, the increase to approximately 60 sr on 20 and 21 September ***is likely associated with*** the hygroscopic growth of smoke particles."

Fourth, several figure cross references were not updated after renumbering. The Table 2 caption cites Figure 6a for the cross sections, now Figure 7a; the text in Sections 5.1.2 and 5.2 cites Figure 3a for the clear sky example, now Figure 4a; and the plume separation text and the Figure 11 caption cite Figure 8a for the purple and pink boxes, now Figure 9a. A careful pass is recommended.

AR: Checked and revised thoroughly.

Please also verify the remaining access dates for consistency.

AR: Checked and updated thoroughly.

A final language edit is also advisable: one residual example is the phrase "approximately 200 Mm^{-1} of across all" in the abstract. In addition, check that relevant acronyms are defined (e.g., HLOS = Horizontal Line-Of-Sight?).

AR: Thanks, revised.

With these corrections I am satisfied that the manuscript is suitable for publication.