Response to Reviewer 1

We thank the reviewer for their detailed and constructive comments.
We have revised the manuscript substantially to address all concerns, including major restructuring of the introduction, significant improvement of figures, clarification of aerosol classification, and expansion of the discussion.
Below are our point-by-point responses. Reviewer comments appear in blue, and our responses follow in black.

General Comments

"The authors present a 10 year data set of lidar and sun photometer observations in Romania. The study is appropriate for ACP. Since it is not a methodology paper it is not appropriate for AMT. It should be moved to ACP after acceptance."

Response: We agree with the reviewer. This is an observational/analysis study, not a methodological paper. Upon acceptance, we will request the transfer of the manuscript from AMT to ACP.

"The results are worthwhile to be published. However, new aspects (new and interesting findings, not known so far) are not presented."

Response: We thank the reviewer for this observation. In the revised manuscript we now emphasise the novelty of the study, namely:
– the first 10-year multi-instrument aerosol climatology for Eastern Europe combining lidar, photometer, and neural-network typing,
– the statistical analysis of the characteristics of the lower-troposphere and high-troposphere aerosol regimes and their seasonal variability in relation to long-range transport
– and the combined interpretation with FLEXPART source attribution.
These points are now stated clearly in the Introduction and Discussion.

"The presentation must be significantly improved."

Response: We have substantially rewritten the manuscript, improving structure, clarity, figures, and discussion depth.

"The figures are partly too small, x-axis and y-axis text and numbers are partly so small that it is impossible to read them in printouts, zoom of 250% is needed at the screen… to study the plots. This is inacceptable!"

Response: All figures have been completely redesigned. Panels, fonts, axes, and symbols are enlarged to meet ACP print standards. No zoom is now required.

"The discussion must be improved, more details are requested. These experts of Eastern European aerosol should be able to deepen the discussion."

Response: The Discussion section has been expanded with the following paragraphs:

Aerosol typing plays a central role in advancing current scientific efforts within EARLINET community (https://www.earlinet.org). Over the last 25 years an extensive number of papers have been published using EARLINET data base (providing aerosol profiling data on a continental scale), and some of them have been focused on aerosol layer classification (e.g. del Águila et al., 2025, Mylonaki et al.,2021, Papagiannopoulos et al.,2018).

Águila et al., 2025 uses machine learning (ML) models for aerosol typing using high-resolution EARLINET data and is currently trained with data from the University of Granada (UGR) station in Spain, which means that it is primarily designed for the specific aerosol types present in this region. A comparative assessment of the Granada and RADO-Bucharest datasets reveals distinct region-specific aerosol signatures. For example, the higher dust depolarization ratios reported in del Águila et al. (～0.25–0.30) contrast with the generally lower values found at our site, reflecting Eastern Europe's less frequent direct dust intrusions and the predominance of mixed or partially aged layers. Our work broadens the geographical coverage of high-resolution aerosol-type climatologies, and offers a foundation for future ML-based classification efforts and for harmonizing lidar-type databases across Europe.

A comparison of the intensive optical properties retrieved in our study with those reported by Mylonaki et al. (2021) reveals both methodological consistency and regional variability across Europe. In their analysis of multiwavelength lidar observations from four EARLINET stations, including Bucharest, Mylonaki et al. reported dust depolarisation ratios typically exceeding 0.25, smoke depolarisation ratios in the range 0.02–0.08, and continental/urban aerosols exhibiting very low depolarisation (<0.05). Our decade-long RADO-Bucharest dataset shows a similar pattern for smoke and continental aerosols; however, dust depolarisation values in our climatology are generally lower than 0.20, reflecting the less frequent and more diluted Saharan dust intrusions that reach south-eastern Romania compared to western and southern Europe. In terms of lidar ratio, Mylonaki et al. (2021) reported dust LR values around 40–50 sr, smoke LR values frequently exceeding 60 sr, and polluted/continental LR values between 45–55 sr. Long-term observations show a dominant LR mode at 48–49 sr across both altitude regimes, with only episodic increases above 70 sr during biomass-burning events—indicating that strongly absorbing smoke layers are less common in the regional transport climatology of Eastern Europe. Similar differences emerge in the Ångström exponent: whereas Mylonaki et al. found AE values for smoke typically >1.5 and for dust <0.5, our dataset displays a bi-modal AE distribution in the free troposphere, with peaks near 1.8 and 0.3, but a mono-modal distribution (median ~0.9) in the lower troposphere driven by mixed and regionally aged aerosols. Together, these contrasts highlight the stronger influence of mixed continental pollution at RADO-Bucharest and the comparatively weaker imprint of "pure" dust and fresh biomass-burning aerosol types relative to the stations analysed by Mylonaki et al. (2021), underscoring the role of regional source regimes and transport pathways in shaping aerosol optical properties across Europe.

Floutsi et al., 2023, presents a data collection (DeLiAn) of intensive optical properties of several aerosol types, as measured by ground based lidars. Apart from campaign measurements, data from four permanent EARLINET stations have significantly contributed to this extensive study. The stations are mainly located in Europe: Leipzig (Germany), Évora (Portugal) and Warsaw (Poland). Our long-term Eastern-European dataset complements the global coverage of DeLiAn, helping to fill a geographical gap (Eastern Europe less sampled) — which is scientifically valuable.

A comparison of the intensive optical properties retrieved in our study with those reported by Ansmann et al. (2021) further supports the interpretation of aged smoke signatures over south-eastern Europe. Ansmann et al. (2021) and references within, found that lidar ratios measured at 355 nm were mostly around 75 ± 25 sr for fresh smoke and 55 ± 20 sr for aged smoke, low to moderate depolarisation ratios (δ ≈ 0.05–0.15), and Ångström exponents typically exceeding 1.5 shortly after emission but decreasing substantially with atmospheric ageing. In our decade-long dataset, aged smoke layers exhibit moderate lidar ratios around 45–55 sr, δ values in the range 0.06–0.10, and an AE distribution with a peak near 1.8 for fresh smoke and near 0.8 for the aged one. These values are consistent with aged, regionally transported smoke but generally fall below the optical extremes observed in major transcontinental plume events analysed by Ansmann et al. (2021). This contrast reflects the different source regimes: while their study captures exceptional long-range transport across hemispheres, our observations document more frequent but less optically intense smoke layers associated with intra-European or Eurasian biomass burning.

"Major revisions are necessary."

Response: Major revisions have been implemented accordingly.

Detailed Comments

Abstract

"The authors write: ….lidar ratios of 48 sr (in the lower troposphere) and 49 sr in the high troposphere suggest varying optical properties …. I do not agree." "Please improve this statement! What did you want to tell?"

Response: This sentence has been corrected and rewritten as follows:

"In the lower troposphere, the extinction-related Ångström exponent shows a narrow mono-modal distribution centered near 0.9, indicating predominantly medium-sized particles, whereas in the high troposphere it becomes bi-modal, reflecting alternating occurrences of small and large particles. Lidar ratio values peak around 48–49 sr in both altitude regions, but their spread is much wider in the lower troposphere—revealing frequent layers of highly absorbing aerosols—while lofted layers in the high troposphere exhibit a narrower range typical of moderately absorbing particles."

Introduction

"The introduction is very long and not straight forward. Instead of 5 pages, two pages are sufficient as an introduction."

Response: The Introduction has been reduced from ~5 pages to ~2 pages and fully rewritten to be concise and focused.

"Concentrate on YOUR contribution in a very compact way! Come to your point, as soon as possible! The reader is not interested in all the details given!"

Response: We agree and have followed this advice. The revised introduction quickly presents the scientific gap, the objectives, and our original contributions.

"Why do you discuss different methods, and especially the negative points here, when you later on just present observational results?"

Response: All unnecessary methodological discussion has been removed from the Introduction.

"Please provide only that information that is needed to understand the results."

Response: Only essential information has been kept.

"The reader wants to know, what is new in this article, and this immediately!"

Response: Novelty is stated explicitly now in the Introduction.

"Long-lasting discussions of methods are not appropriate."

Response: These discussions have been removed entirely.

Citation Formatting

"P 4, line130: Reference options (TEX) are \citet and \citep, you always use \citep , but in line 130 you should, e.g., \citet{Amiridis….}. There are many places where \citet is needed, please improve!"

Response: All citations have been checked and corrected. We now use \citet where required, consistent with ACP style.

Methods / Background Sections

"P 4, lines 138 to 170: To repeat it again here: why is all the information regarding methods and techniques given? The topic is the presentation of aerosol observations in Romania and discussion of findings! So, please concentrate on that, only!"

Response: The entire section has been removed or condensed. Only minimal method descriptions necessary to interpret results is included now.

"P 5, line 177: What does RADO mean?"

Response: We now define RADO at its first use: *Romanian Atmospheric 3D Observatory*.

Section 2

"Again, keep the description short, please!"

Response: Section 2 has been shortened significantly.

"The reader is interested in new aspects, not in all these general points and descriptions listed and presented."

Response: General descriptions have been removed.

"A compact introduction into the complex aerosol conditions in Europe is needed, followed by a specific focus on Eastern Europe."

Response:

We have rewritten the Introduction addressing also Europe-wide aerosol context and Eastern European specifics, highlighting that across Europe, aerosol conditions are shaped by the coexistence of several major source regimes, including persistent anthropogenic pollution, recurring Saharan dust intrusions, marine inflow from the Atlantic and regional seas, and seasonal biomass-burning plumes.

This combination produces strong spatial gradients, frequent vertical layering, and extensive mixing that challenge both model representation and remote-sensing retrievals. Within this broader context, Eastern Europe occupies a particularly complex position, as it lies at the intersection of continental pollution from Central and Eastern Europe, dust transport from North Africa and occasionally the Middle East, marine influence from the Black Sea, and smoke from both regional and remote fires. These overlapping transport pathways and the large variability of synoptic conditions make Eastern Europe a key region where long-term, vertically resolved ACTRIS observations are essential for documenting the evolution, mixing state, and radiative impact of aerosols.

"This is the role of an Introduction section… introduce into the topic, and what the gaps in our knowledge are and how this study contributes to fill the gap."

Response: The Introduction has been rewritten accordingly.

"Most of the information in the first 7 pages is not needed and should be removed."

Response: It has been removed.

"A short introduction and a straight forward description of used technique and data analysis methods can be done within 3 pages."

Response: The introduction + methods description now fits within approximately 3 pages.

Section 3 – Results and Discussion

"All figures need to be improved! AMT/ACP standard is to have (a), (b), (c), …. in all panels of a given figure."

Response: All figures have been redesigned with proper panel labels.

"Most of the figures are too small… These small figures are inacceptable!"

Response: All figures have been enlarged and reformatted.

"What do we learn from Figure 1?"

Response: The initial figure has been removed and replaced with a monthly time series of the mean and median values of Aerosol Optical Depth, Fine Mode Fraction, and the percentage of Angstrom Exponent values greater than 1. This way, the figure effectively reveals several key aspects: the low-AOD climatology; the strong long-term stability of the fine-mode fraction; and the predominance of fine-mode particles.

"What do we learn from Figure 2?"

Response: Figure 2 has been replaced with seasonal violin plots of the three variables. The width of the violin directly corresponds to the smoothed probability density function, or the histogram equivalent. This method allows for a more detailed comparison of the shape, skewness, and modality of the seasonal distributions across all years. The median and the interquartile range are visually marked by horizontal lines within each violin.

“Figure 3 is confusing!”

Response: Figure 3 has been reduced to the aerosol typing graph based on the joint probability density of AOD and AE. The main 2D colour plot shows the relationship between AOD (x-axis) and AE (y-axis). Each of the coloured regions describes a different dominant aerosol regime. The darker the color of a region the higher the density of the points composing said region. The figure is made by incorporating the thresholds established by D'Almeida et al. (1991), Dubovik et al. (2002), Toledano et al. (2007). The full thresholds have been added as a supplement to the article.

“The aerosol types need to be defined! Maybe I missed it!”

Response: We added a complete table of definitions for all aerosol types characterised by the sun-photometer measurements in the supplement.

“In Figure 4, the increasing size of the circles in the plot obviously indicates the year! But this is not shown in the legend.”

Response: Figure 4 has been replaced with a heatmap of the most statistically dominant aerosol type calculated for each month and year.

“What is now the message of this colorful plot?”

Response: Figure 4 has been replaced. The new figure is explained in the newly written text.

“Marine particles so often dominate in polluted Romania … I am confused.”

Response: We agree that the presence of Marine-dominant days in a continental region requires clarification. In the revised manuscript, we added an explicit explanation noting that the “Marine” category in our optical classification scheme represents a coarse-mode optical signature, not necessarily pure sea-salt or true marine air masses. This clarification has also been added to the table to explain each of the dominant aerosol types, which has been added to the supplement.

“Figure 5 … what is different here, what is new?”

Response: The original Figure 5 has now been replaced with a heatmap showcasing the dominant aerosol type for each recorded day over the entire measurement period. The new figure is explained within the text.

Section 3.2

1. ***“However, I personally do not trust much in automated solutions of lidar inversion methods (ill posed problem). Especially the backscatter coefficient at 1064 nm has a sensitive impact on the inversion product, but it is always given with high uncertainty. The Rayleigh calibration at 1064 nm remains always a problem.***

Response: We fully acknowledge the limitations and ill-posed nature of lidar inversion, particularly at 1064 nm where Rayleigh calibration remains challenging and retrieval uncertainties are inherently higher. Although SCC-(Single Calculus Chain) derived products were used as input for the NATALI algorithm, all datasets underwent systematic manual inspection prior to aerosol typing. Additional quality-control tools were applied to verify signal integrity, calibration stability, and consistency across channels before submission to the ACTRIS database. Furthermore, the RALI system is regularly evaluated within the CARS (Center for Aerosol Remote Sensing) QA (Quality Assurance) framework, which includes alignment optimization, calibration checks, and refinement of the SCC configuration to minimize uncertainties in the inversion products. These procedures are essential for generating reliable aerosol typing results, and ACTRIS maintains continuous efforts to refine lidar processing workflows and reduce uncertainties in the products used for regional and continental aerosol studies.

"Figure 6: The y-axis has no scale."

Response: A full axis scale has been clearly marked.

"Everything in the figures should be explained."

Response: Symbols, lines, and distributions are now explained in detail.

"Text is so small, why?"

Response: Font sizes have been increased.

"Figure 7: Everything is simply too small… What is the message here?"

Response: The figure has been enlarged, reformatted, and its message clearly described.

"P 13, lines 423-430. These are general statements and could be done even without any observation! Now we have a zoo of aerosol types: continental smoke, mixed smoke, mixed dust, dust polluted, and so on. What do we learn from the presented results? Is there any specific aspect of Eastern European aerosol compared to other European aerosols."

Response: A paragraph has been added to underline the specificity of the Eastern European aerosol compared to other European aerosols as follows:

"Our long term analysis shows that aerosol layers over South East Romania are rarely composed of a single dominant type and are instead characterized by frequent mixtures of continental, smoke, and dust components. This persistent mixing reflects the geographical position of the region, which is influenced simultaneously by local anthropogenic emissions, agricultural burning, and episodic transport from the Sahara and Eastern Europe. Compared to other European locations, the RADO-Bucharest site shows a higher prevalence of mixed continental–smoke layers in the lower troposphere and more frequent dust–smoke combinations at mid altitudes, indicating that biomass burning plumes arriving from Eastern Europe often interact with advected mineral dust. These mixed signatures are well captured by NATALI, with high resolution classifications ~~clearly~~ identifying combinations such as dust polluted, continental smoke, and mixed dust, which are not as frequent at sites dominated by either dust (southern Europe) or pollution (central and western Europe) alone. The difficulty in distinguishing continental polluted from smoke in low resolution also reflects the regional dominance of fine mode combustion aerosols, which share similar spectral properties. Overall, the classification patterns reveal that Eastern European aerosol at this site is defined not by a single type but by recurrent mixtures driven by simultaneous influence of local emissions, agricultural fires, and intermittent dust transport."

"Figure 8: Again, this figure is much too small."

Response: The figure has been enlarged and redesigned.

"Figure 9: continental aerosol and continental polluted aerosol, is there a difference?"

The detailed description of each aerosol type is provided in Nicolae et al. (2018), Tables 1 and 2. In brief, continental aerosols (land-origin) are represented by mixtures of water-soluble particles, insoluble material, and soot, whereas continental polluted aerosols (typically associated with industrial regions) include the same components with an additional sulfate fraction.

To clarify this distinction and the broader NATALI aerosol-typing framework, we have added a dedicated explanatory section in the revised manuscript, as follows:

"The main aerosol category is determined across the six possible outputs, each representing a dominant type that may in-clude up to 50 percent presence of other components as minor traces: continental, continental polluted, smoke, dust, ma-rine, and mineral mixtures. In high-resolution mode, NATALI further refines these classes into a set of fourteen advanced aerosol types that capture both pure and mixed states. These include: Continental (water soluble, insoluble, soot), Dust (mineral nucleation, mineral accumulation, mineral coarse mode, water soluble, soot), Continental Polluted (water soluble, soot, insoluble, sulfate), Marine (water soluble, sea salt accumulation mode, sea salt coarse mode, soot), Smoke (water solu-ble, soot, sulfate), Volcanic (mineral nucleation, mineral accumulation, mineral coarse mode, water soluble, sulfate), as well as mixed and binary types such as ContinentalDust (continental + dust), MarineMineral (dust + marine), ContinentalSmoke (continental + smoke), DustPolluted (dust + smoke or dust + continental polluted), Coastal (continental + marine), Coastal-Polluted (continental polluted + marine), MixedDust (continental + dust + marine), and MixedSmoke (continental + smoke + marine). The correspondence between the aerosol types retrieved by NATALI in high- and low-resolution modes is presented in the results section."

"Figure 10 seems to be interesting, but a discussion is not given."

Response: Thank you for highlighting this issue. The discussion for this figure was inadvertently omitted from the original manuscript. In the revised version, we have included a comprehensive interpretation of all panels as follows:

"Long-range transported biomass burning aerosols (smoke) and their variation with atmospheric evolution (ageing) have been extensively studied during the last years using lidar measurements. To distinguish between fresh and aged smoke we have been using the ratio of lidar (extinction-to-backscatter) ratios (LR532/LR355). It has been observed that this changes rapidly from values <1 for fresh to >1 for aged particles (Nicolae et al, 2013).

Figure 10 presents the distribution, central tendency, and spread of several lidar-derived intensive optical properties used to characterise fresh and aged smoke: (a) altitude, (b) linear particle depolarisation ratio at 532 nm, (c) extinction-related Ångström exponent (AE), and (d) lidar ratio (LR) at 355 nm. Together, these panels provide a consistent physical picture linking smoke ageing to transport altitude, aerosol morphology, and optical signatures.

Figure 10 (panel a) illustrates seasonal differences in the vertical distribution of fresh biomass burning aerosol layers. During spring, the detected layers extend to substantially higher altitudes, reaching up to 2 km, while in summer and autumn the maximum heights are noticeably lower. Despite this contrast in upper extent, the median layer height remains close to 1 km for all three seasons. For winter, fresh smoke cases are largely absent, most likely because unfavourable weather conditions (persistent low clouds and overcast skies) frequently prevented lidar observations, rather than because such aerosol events did not occur.
For aged smoke particles, the vertical distribution shows a contrast between winter and the other seasons. During

winter, the aged aerosol layers remain more confined, typically not exceeding 2 km, whereas in spring, summer, and autumn the layers extend higher, reaching 3 km and up to about 3.5 km in summer. This pattern suggests that in winter the planetary boundary layer constrains the vertical mixing of aged smoke, keeping the aerosol trapped in the lower part of the atmosphere, while in the warmer seasons deeper boundary layers allow the layers to ascend to higher altitudes.

Figure 10 (panel b) presents the distribution of fresh and aged smoke particles as a function of the particle depolarization ratio at 532 nm. For the RADO-Bucharest site, aged smoke typically exhibits depolarization values between 0 and 15 percent, with only a few isolated cases exceeding this range. Fresh smoke shows a narrower distribution, with most values constrained between 5 and 15 percent. This behaviour is consistent with the expected microphysical evolution of biomass burning aerosols, where aging processes generally reduce particle asphericity and broaden the variability of the depolarization signal.

Figure 10 (panel c) presents the distribution of fresh and aged smoke particles as a function of the 355 nm lidar ratio, revealing a bimodal behaviour for both aerosol types. Fresh smoke shows two distinct clusters, with LR values predominantly occurring in the range 60 to 80 sr and a second group above 100 sr. A similar pattern is observed for aged smoke, although the lower LR cluster shifts toward smaller values, approximately 27 to 60 sr, while the upper cluster remains above about 75 sr. The presence of these two clusters for both fresh and aged cases suggests the influence of at least two major smoke sources affecting the region, each characterized by distinct optical properties."

Section 3.3

"The Flexpart simulation shows the expected result. Maybe a bit unexpected that Europe is so dominating."

Response: The results are not unexpectedly large in terms of the contribution from European sources, since the MARS station is positioned close to Bucharest (~6km of its periphery) in southeastern Romania, on the eastern part of Europe. Due to the location-specific climate (Bogdan, O. and Niculescu, 2005), air masses, in their circulation (most frequently westerly circulation) remain longer over large regions with aerosol sources from Europe before reaching the measurements' site.

"I do not understand the colors of the bars."

Response: The color scheme is now clearly defined in the text. For clarity, the color bars in Figure 11 have been described extensively in the text and tables with source distribution (%) have been introduced in the supplement.

"At the end I miss a comparison with other EARLINET studies."

Response: A comparison with other EARLINET findings has been added to the Discussion.

Final Remarks

All reviewer comments have been addressed in detail.
We believe the revised manuscript is significantly improved and meets ACP expectations.

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