• Response:

We sincerely thank the reviewer for the comprehensive evaluation of our manuscript and the constructive feedback. The suggestions raised have led to substantial improvements in both scientific content and presentation. We have revised the manuscript extensively, addressing all structural, methodological, and interpretational issues highlighted in the review. A detailed, itemized response to each comment is provided in the following sections.

Reviewer comments appear in blue, and our responses follow in black.

General assessment

> ***"The paper examines aerosol characterisation at the RADO-Bucharest station in Romania, part of ACTRIS. The authors use sun/sky/lunar photometer and lidar measurements, combined with the NATALI neural network, to distinguish between aerosol types in the lower troposphere and above the boundary layer, and FLEXPART retro-plume analysis to assess potential sources."***

Response: We thank the reviewer for summarising the study.

> ***"However, the manuscript is difficult to follow, reads more like a descriptive study or technical report than a scientific paper, and requires major revision."***

Response: The manuscript has undergone extensive restructuring. The introduction and methodology were shortened and refocused, and the results/discussion sections were rewritten to emphasise scientific interpretation rather than descriptive content.

Introduction

> ***"The introduction is excessively long, ending with only a single sentence that addresses the paper's objective."***

Response: The introduction has been substantially shortened and rewritten. The objective is now stated clearly and earlier, and the final paragraph provides a structured transition to the methodology.

> ***"Lines 57–73, which provide basic definitions of aerosol properties, should be removed or drastically shortened, as this information is already well known to the community."***

Response: These lines have been removed.

> ***"Up to line 137, the text remains overly descriptive and does not help the reader understand the actual objective of the study."***

Response: All overly descriptive content has been removed or rewritten. The introduction now focuses directly on the scientific motivation and research questions.

> ***"The introduction also ends abruptly, with no clear transition to the methodology or explanation of how the objective will be achieved. Leaving such information entirely to the methodology section is not acceptable."***

Response: We added a transitional paragraph at the end of the introduction explaining the approach, the dataset, and how the objective is addressed.

“How are the ACTRIS-CARS protocols relevant to the objectives of the paper, and why were they included?”

Response: We now explicitly explain that ACTRIS–CARS protocols ensure measurement traceability and comparability across long-term datasets. This justification is added and the section shortened.

“Likewise, the extensive discussion of GARRLiC/GRASP is misleading, since these methods are not applied in the study.”

Response: The discussion of GARRLiC/GRASP has been removed.

“As written, the introduction gives the impression that the analysis will rely on a synergy of photometer and lidar measurements processed with GRASP, which is not the case.”

Response: We revised the introduction to remove this unintended implication.

Methodology

“The methodology section is quite lengthy and lacks subchapters dedicated to the different instruments or methods used, which would help make it more accessible and easier to follow.”

Response: The methodology has been reorganised into clear sub-sections (site, lidar, photometer, NATALI, FLEXPART).

“Why is so much space devoted to describing the instruments and retrieval methods? Photometers and lidars are well-established, and their detailed characterisation has already been extensively documented in the literature. This level of detail is unnecessary here. You should limit the description to the basics and only add specific information if there are instrumental modifications unique to the RADO-Bucharest site.”

Response: We shortened the entire instrument description, keeping only site-specific details.

“Regarding FLEXPART, the resolution of the meteorological input data is not specified and should be clearly stated. Furthermore, given that this paper is submitted to an ACP/AMT special issue, the description of the model setup is far too superficial.”

Response: The description of the FLEXPART model configuration, as well as the necessary references, have been added to the text in methodology section.

“Details on the so-called ‘unique turbulence model’, the wet deposition scheme, and the parameterisation of gravitational settling are missing and need to be explicitly described.”

Response: A brief description of these improvements brought to FLEXPART version 10.4 as well as the related references has been added to the text. Details about the improvements made to FLEXPART 10.4 (unique turbulence model, wet deposition schema and the parameterisation of gravitational settling) are given in reference Pisso et al., 2019. In our study FLEXPART was set to use these improvements.

“Lines 290–295 could be removed.”

Response: These lines have been removed.

Figures

> ***"A general issue in the results section is the poor quality of the figures. All figures require improvement, with clearer scales, larger fonts, and properly labelled axes."***

Response: All figures were re-drawn with larger fonts, improved scales, and clear axis labels.

> ***"All figures and captions should be improved with the use of a), b), c) etc. instead of 'left panel', etc."***

Response: All multi-panel figures now use (a), (b), (c), etc. consistently.

> ***"In the text as well, when discussing the figures, they should be in line with the ACP/AMT requirements of Fig. a) in the text, or Figure a) when at the beginning of the sentence."***

Response: All figure references in the text were updated accordingly.

Aerosol classification

> ***"The use of the term 'polluted' as an aerosol category is imprecise and should be avoided; more appropriate terminology would be urban/industrial or anthropogenic."***

Response: We replaced "polluted" with "urban/industrial"

> ***"Your use of continental is left to interpretation."***

Response:

Our classification scheme, based on the joint analysis of AOD and AE is designed to differentiate between the dominant optical signatures present in our region. We explicitly use the following approach to prevent ambiguity:

- The urban/industrial cluster is reserved for the highest AE values, which are indicative of small, fresh particles. In the context of the OPAC model, this classification optically corresponds closest to the Continental Polluted type.
- The continental type is explicitly defined by the optical properties: AE ~1.0-1.3 and low to moderate AOD. This signature represents the regional background aerosols, which, at least for the region of the site, is an inevitable mixture of aged natural and fine-mode anthropogenic aerosols that have spread across a large area. By setting this AE range, we distinguish it from the urban/industrial type and the dust/marine types. This category, therefore, optically acts as a regional proxy for the continental mixed or continental clean categories in the OPAC framework.

We will add to the supplement a dedicated table with definitions like this:

Table 1: Detailed description of dominant aerosol types

Aerosol Type	Angstrom Exponent Threshold	Aerosol Optical Depth Threshold	Physical interpretation	Notes
Urban/Industrial	AE > 1.52	0.2 < AOD < 0.4	Dominated by fine anthropogenic particles such as combustion emissions, traffic/industrial sources, and secondary aerosols	Most common in urbanized or industrial regions; often dominating during stagnant winter conditions and temperature inversions
Continental	AE >1.2	AOD < 0.2	Aged, regionally transported fine-mode aerosols originating from mixed anthropogenic and biogenic sources	Represents the persistent baseline background in most continental regions
Dust	AE < 1.15	AOD > 0.2	Mineral dust, typically from Saharan or local resuspension processes	AE for "pure dust" can fall <0.5 near source, but transported dust often shows higher AE due to mixing with fine-mode particles
Marine	AE < 1.2	AOD < 0.2	Optically coarse-mode particles resembling maritime aerosols (sea salt)	In continental regions, this class often reflects generic coarse-mode aerosols (weak dust, humidified particles), not necessarily true marine air masses
Mixed	1.15 < AE < 1.52	AOD > 0.2	*Overlapping contributions of both fine and coarse modes; optically heterogeneous column*	Common during transitions, high-AOD events, or multilayer structures (e.g., dust above pollution)
Biomass Burning	AE > 1.52	AOD > 0.4	*Fine-mode smoke from wildfires, agricultural burning, or regional biomass burning episodes*	More common in late summer/early autumn depending on region; may overlap with Continental fine-mode regime

“Using continental without clarification introduces confusion and must be explicitly defined in your classification.”

Response: A dedicated table was added to the supplement and now provides clear definitions (see previous table).

“What is Tight Continental?”

Response: The term “Tight Continental” has been removed.

“Weighting annual aerosol fractions by the number of measurements per month or year is not recommended… A better approach is to use equal-month or equal-season weighting… and to report 95% confidence intervals.”

Response:

Equal-moth or equal-season weighting, along with confidence intervals, is the appropriate methodology for deriving unbiased long-term annual aerosol statistics. We fully acknowledge that this approach is superior for calculating robust mean annual fractions. However, the purpose of the heatmaps differs from such statistical objectives. The new Figure 4, which existed in the original text as well, is designed to show the dominant aerosol type within each month, pointing out temporal shifts and seasonal patterns rather than producing a weighted annual mean.

Figure 5, which replaced the old Figure 4, serves as a high-resolution, descriptive visualisation for identifying episodic events and illustrating the persistence of the Continental background regime. Because the figure is used for qualitative event detection rather than long-term statistical averaging, applying monthly or seasonal weighting is not necessary for the stated purpose.

“Additionally, include a table of N per month/season/year so readers can evaluate the support for each estimate.”

Response: This table has been added to the Supplement as follows:

Table 2: Monthly count of dominant aerosol types (the number represents the total number of points classified as a certain aerosol type)

Month	Biomass	Continental	Dust	Marine	Mixed	Urban/Industrial
1	9	3885	222	372	246	441
2	74	3234	622	396	239	775
3	492	3061	449	351	574	1723
4	143	4612	911	1100	666	1933
5	48	4550	911	1629	1208	970
6	333	3141	1391	404	1354	3362
7	1849	5415	1900	278	1731	6522
8	1646	5945	771	432	3098	6806
9	433	4451	747	1339	1693	2634
10	281	5115	544	1423	617	1492
11	55	2903	225	978	369	292
12	6	2293	14	384	105	349

Table 3: Seasonal count of dominant aerosol types (the number represents the total number of points classified as a certain aerosol type)

Season	Biomass	Continental	Dust	Marine	Mixed	Urban/Industrial
DJF	89	9412	858	1152	590	1565
JJA	3828	14501	4062	1114	6183	16690
MAM	683	12223	2271	3080	2448	4626
SON	769	12469	1516	3740	2679	4418

Table 3: Yearly count of dominant aerosol types (the number represents the total number of points classified as a certain aerosol type)

Year	Biomass	Continental	Dust	Marine	Mixed	Urban/Industrial
2015	679	1820	194	169	343	1507
2016	617	3133	815	352	607	2406
2017	509	4125	802	310	962	2182
2018	415	1726	765	738	1250	1608
2019	460	3940	180	644	1873	3252
2020	731	10794	912	1234	956	5161
2021	831	8802	2675	1354	2336	4870
2022	550	8108	1203	1526	1672	3381
2023	577	4963	951	1810	1610	2669
2024	0	1194	210	949	291	263

Specific interpretation issues

"I cannot identify any extended Marine event in 2021 from the figure. Could you clarify what this conclusion is based on? Did you mean 2024?"

Response: Thank you. This was an error. The 2021 statement has been removed and corrected in the text with 2024.

"For clarification, you remove the HT cases when there's no layer below the PBL (because you mention 'all the layers that are located above the first layer'?)"

Response: We do consider all HT cases. If there is no layer in the LT, any layer above 3km is classified as HT case. We added a clarification of the layer-selection procedure in section 3.2.

"In the interpretation of Figure 6, there is an inconsistency in the statistics: you discuss the median for LT but the mean for HT; please clarify which metric is being used."

Response: We now use the median for both LT and HT, and this is stated clearly.

"The reported lidar ratios of 48 vs. 49 do not constitute a meaningful separation… the statement that these values 'suggest varying optical properties' is an overstatement."

Response: We revised this statement in the abstract and main text 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."

"I can understand dust and smoke as predominant types in the HT, but what about the continental clean? The reported ~50% fraction appears unexpectedly high. Please clarify whether this reflects actual aerosol composition, or if it could result from limitations of the classification algorithm, low-concentration background aerosols, or misclassification of mixed layers. Neural networks like NATALI or other aerosol typing algorithms can sometimes misclassify mixed or low-concentration aerosols as 'continental clean', especially at high altitudes where signal-to-noise is lower"

Response:

The reported ~50% fraction reported for the predominant aerosol type as continental it reflects the actual aerosol composition: when the layer is classified as continental it means that at least 50% of the composition of the layer is continental; It is not related to low concentrations and it is not influenced by the low signal-to-noise at high altitudes. We added the following phrase in the methodology section of NATALI algorithm to clarify better this aspect:

"The main aerosol category is determined across the six possible outputs, each representing a dominant type that may include up to 50 percent presence of other components as minor traces: continental, continental polluted, smoke, dust, marine, and mineral mixtures."

"Figure 9 requires improvement. You refer to frequency, but no actual numbers are provided; the current scale leaves too much open to interpretation. Please include counts or percentages to make the data interpretable."

Response: The figure has been redesigned with explicit percentages and counts.

"Figure 10 is presented without a description or interpretation. It is not the reader's task to disentangle the meaning of the plots. The explanation should clarify how depolarisation is used to differentiate between aged and fresh smoke, and this should be introduced first. Only after that should you discuss the role of altitude, Ångström exponent, and lidar ratio."

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."

FLEXPART analysis

"The current Section 3.3 is too brief and largely descriptive. Currently, it only paraphrases what is already visible in Fig. 11 and does not sufficiently leverage the potential of the FLEXPART simulations. As a result, the section lacks depth, fails to connect with the observational findings, and does not convincingly demonstrate the added value of FLEXPART in the study. To improve the scientific quality and readability of this section, I recommend the following mandatory revisions:"

Response: Section 3.3 has been revised according to the recommendations

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"Please provide quantitative results (e.g., mean seasonal percentage contribution ± standard deviation). A summary table in the Supplement would also be very useful."

Response: Quantitative values have been added, and a summary table is included in the Supplement.

"The choice to split retroplume clusters at 2 km altitude is arbitrary and inconsistent with your own discussion of the PBL, where a climatological mean of ~1300 m was already established. If you intend to use 2 km as a threshold, this requires justification. For instance: o Why is 2 km

chosen rather than the mean PBL height (1.3 km)? o Does 2 km correspond to a standard practice in FLEXPART studies? Please cite."

Response: As recommended, for consistency in the PBL discussions, we changed the threshold value to 1300 m, which represents the climatological mean of the PBL. The FLEXPART analysis was revised accordingly.

Lines 469-471 changed with: "To distinguish between the influence of local and long-range transport in the FLEXPART retro-plumes analysis, the retro-plumes trajectories (corresponding to altitude levels from 500 m to 8.0 km) were split into the two clusters (LT and HT) using the threshold value of 1.3 km, altitude corresponding to the climatological mean value of the PBL."

Added to the text: "This value was selected to ensure consistency in the analysis of data obtained from lidar and photometer measurements with data obtained from FLEXPART. Moreover, this threshold value is high enough to avoid surface friction and terrain/topography effects in the area where the RADO Bucharest station is located, but low enough to capture important meteorological patterns such as the presence of low-level clouds, temperature variations, wind changes and moisture transport.".

"The definition of source regions is not clear. Please show a map with the spatial masks used for Europe, Sahara, North Africa, etc., so that the classification is reproducible."

Response: A map with the spatial masks used for Europe, Sahara, North Africa, North America, Middle East and Siberia was added in the manuscript as Figure 11.

Lines 473-475 changed with: "1. Sources that contribute to the aerosol budget at a given location and are distributed in a single region. In this category, following sources were defined: Europe (continent), North Africa (Algeria, Egypt, Libya, Morocco, Sudan and Tunisia), Sahara (West Sahara, Mali, Niger, Chad and Mauritania), Middle East (Arabic Peninsula, Iran and Iraq), North America (Canada and USA), Siberia."

Added to the text the figure with the distribution of aerosol sources regions used in the FLEXPART analysis, as Fig. 11

Line 478 changed with: "The distribution of regions with aerosol sources used in the FLEXPART analysis is shown in Fig. 11 and the distribution of sources by season and by the two clusters is shown in the Fig. 12."

Lines 479 – 481: Replaced Fig. 11 with Fig. 12.

"The methodology section describes improvements in FLEXPART physics (turbulence, wet deposition, ERA5 input), but none of these are discussed in the results."

Response: The description of the improvements in FLEXPART and ERA5 input data, as well as the necessary references, have been added to the text, in methodology section.

"How did including wet deposition or the new turbulence scheme affect their retroplumes compared to earlier FLEXPART studies? "

Response: Comparisons between the improvements brought in the FLEXPART versions 10.4 with other FLEXPART versions were not the subject of our study. The comparative study between the results obtained with different versions of the FLEXPART model could be the subject of a future paper.

"FLEXPART analyses cannot be interpreted in isolation. You should discuss the prevailing synoptic conditions (e.g., seasonal circulation, anticyclonic vs cyclonic regimes, vertical transport patterns) that explain the seasonal differences in source contributions."

Response: A brief description of the synoptic conditions specific to Romania has been added to the text in section 3.3.

"At present, FLEXPART results are disconnected from the lidar/photometer data. Please explicitly connect transport simulations with the observed aerosol types."

Response: The connections between transport simulations and the types of aerosols observed in the lidar data have been included and described explicitly in the text. The variation in the optical properties of dust and smog depending on the sources is presented in the text, in Tables 1 and 2.

"Unless you address these issues… this section… should be removed."

Response: Section 3.3 has been fully revised and retained.

Conclusions

"The final paragraph from Chapter 4 is overly general and does not provide a critical perspective. Please expand it to include a discussion of the broader implications of your findings, potential limitations of your study, and directions for future work. For example, how could this work inform improved aerosol modelling, observational networks, or policy-relevant assessments of air quality and climate?'

Response: The final paragraph was rewritten to include implications, limitations, and directions for future work as follows:

Beyond the descriptive patterns described above, the results have several implications for how aerosol variability should be represented and observed in this region. The frequent occurrence of mixed aerosol states, in particular the recurring continental-smoke and dust-smoke combinations identified by NATALI, points to a need for regional chemical transport models to better resolve vertically structured mixing and the seasonally varying ageing of fine-mode particles. The extended record also demonstrates the importance of maintaining ACTRIS-grade, co-located lidar and photometer observations at sites where multiple transport pathways intersect, since column-only products struggle to capture the layered structure revealed here. At the same time, the analysis is constrained by reduced lidar sampling during cloudy and winter periods, the limited capability of the low-resolution NATALI mode to separate fine-mode types with similar spectral behaviour, and the relatively small set of layers with calibrated depolarisation for the high-resolution classification. Addressing these limitations will require expanding the multi-parameter retrieval capability toward combined lidar-radiometer inversions and integrating these with regional modelling tools such as FLEXPART-CTM coupling. Such developments would allow a more quantitative assessment of source contributions and radiative effects, and would strengthen the use of long-term ACTRIS datasets for evaluating emission-reduction policies and constraining aerosol-climate interactions in Eastern Europe.

Specific comments

“• Revise your citations. When citing, correct is ‘Nicolae et al. (2023) have found …’ not ‘(Nicolae et al., 2023) have found…’. E.g. lines 130, 165, 213 etc Improve the abstract.”

Response: Citations have been corrected and the abstract has been improved.

“Improve all figures.”

Response: All figures have been improved as requested.

“Line 93: ‘from the’”

Response: Corrected.

“Line 228: cite the OPAC database”

Response: Citation added.

Hess, M., P. Koepke, and I. Schult, 1998: Optical Properties of Aerosols and Clouds: The Software Package OPAC. Bull. Amer. Meteor. Soc., 79, 831–844, https://doi.org/10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2.

“Line 244: review the aerosol type classification.”

Response: The classification scheme as is described in details in Nicolae et al,2018) has been fully reviewed and written also in this paper as follows:

“The main aerosol category is determined across six possible outputs, each representing a dominant type that may include up to 50 percent presence of other components as minor traces: continental, continental polluted, smoke, dust, marine, 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 soluble, 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), CoastalPolluted (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.”

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