ACP manuscript egusphere-2026-998 - Enhancement of ammonium nitrate aerosol in the Northern Hemisphere lower stratosphere linked to Asian summer monsoon outflow

Ekinci et al., https://doi.org/10.5194/egusphere-2026-998

We would like to thank **Reviewer #2** (https://doi.org/10.5194/egusphere-2026-998-RC2) for the detailed comments and constructive suggestions, which encouraged us to critically revisit the manuscript and helped us improve its clarity and overall presentation.

The responses are structured according to the following format:
Comments by the reviewer
Responses by the authors
Corresponding changes made in the manuscript; In addition, everything that was removed has been crossed out. For the sake of clarity, individual words that were changed have also been highlighted in green.

General points:

1. **The analysis of this paper is based on 6 research flights (3 for the early and late phase each). I am missing a discussion how representative this data base is for the conclusions drawn in this paper. Figure 4 indicates that there are data gaps for certain potential temperature and equivalent latitude ranges, but I think this should be discussed how this affects the conclusions.**

   Thank you for this important comment. We agree that the representativeness of the dataset needs to be discussed more explicitly. The analysis is based on six research flights, with three flights representing the Early Phase and three flights representing the Late Phase. Therefore, the dataset should not be interpreted as a climatological representation of the entire Northern Hemisphere ExLS. Instead, it represents the ExLS background air masses sampled during the PHILEAS campaign in August and September 2023. While the PHILEAS observations themselves are not intended to represent a climatology of the entire Northern Hemisphere ExLS, the ASM is expected to have a large influence on the ExLS as shown in earlier studies (e.g. Yu et al., 2017; Fadnavis et al., 2024; Graßl et al., 2024).

   The main conclusion of an enhanced ammonium nitrate signal in the sampled ExLS background air during the Late Phase is supported by several independent lines of evidence. These include the ERICA-AMS bulk aerosol composition, the ERICA-LAMS single particle analysis, the CLaMS surface origin tracers and age spectra, and the trace gas observations. These independent diagnoses are consistent with each other, which reinforces the confidence that the change from summer to autumn observed in Fig. 4 is not solely due to the absence of measurements/samples in certain areas.

   However, the conclusions are now more explicitly restricted to the sampled ExLS background air masses and to the dynamically and geographically covered regions of the campaign. The data gaps in Fig. 4 indicate that not all potential temperature and equivalent latitude ranges were sampled equally in both phases. Therefore, we do not interpret unsampled or poorly sampled bins (<10 data points) as representative of the broader ExLS.

We have added a discussion of these limitations and clarified that the data gaps primarily affect the comparability of a few equivalent latitudes versus potential temperature ranges, while the qualitative phase difference in the sampled air masses remains supported by the combined observational and model evidence.

L241: *"[...] Second, we demonstrate that the ExLS background composition in the Early Phase of PHILEAS between 45°N and 65°N was characterized by aerosol that could be associated with the ASM region. [...]"*

L269: *"[...] Furthermore, it is important to consider that the interpretation of the comparison between the Early Phase and the Late Phase is limited by the sampling characteristics of an aircraft-based field campaign. The results and interpretation correspond and are limited to air masses sampled during the PHILEAS flights, rather than corresponding to the entire Northern hemisphere ExLS region. The analysis is based on six research flights, three of which were assigned to each phase, and therefore does not represent a climatological description of the entire Northern Hemisphere ExLS. While the PHILEAS observations themselves are not intended to represent a climatology of the entire Northern Hemisphere ExLS, the ASM is expected to have a large influence on the ExLS as shown in earlier studies (e.g. Yu et al., 2017; Fadnavis et al., 2024; Graßl et al., 2024). The measurement data, however, provide a sufficient basis for characterizing the sampled background air masses, particularly in the range between 45° and 65° N and above 360K. At the same time, it must be taken into consideration that the air masses examined do not cover the equivalent latitudes and potential temperature ranges equally in both phases. Therefore, bins with insufficient data coverage (fewer than 10 data points) in Fig. 4 and 5 are considered as not statistically significant. Bins that were not equally covered in the Early and Late Phases were also excluded from the calculation of differences. Therefore, the conclusions derived from this analysis refer to the ExLS background air masses investigated within the scope of this campaign. [...]"*

L434: *"[...] Although data gaps limit the spatial completeness of the comparison in ExLS, they do not significantly affect the qualitative conclusion, as both in-situ measurements and model data support the results. [...]"*

2. **For the analyses, particle fractions were plotted and discussed. I wonder if the increase or decrease of these PFs are also reflected in an increase or decrease of absolute particle numbers. Or is the PF even the fraction of those particles, which can be detected by ERICA? Then absolute numbers would be even more interesting.**

We thank the reviewer for this helpful comment. The particle fraction (PF) is defined as the number of particles assigned to a specific ERICA-LAMS particle type, divided by the total number of ERICA-LAMS particles detected and classified within the respective potential temperature and equivalent latitude bin. Therefore, the PF represents the proportion of a given particle type within the total ERICA-LAMS particle population, rather than the proportion of all atmospheric particles. The particle concentration of aerosol particles is measured by the first detection stage (N_0).

We agree that the particle number concentration provide additional information. Following the approach of Appel et al. (2022), we calculated the scaled number concentration of each particle type for each bin by multiplying the PF with the respective particle number concentration (as N_0). This analysis shows that the key findings derived from the PF-based vertical profiles are also reflected in the corresponding profiles when the number concentration is taken into account.

To make this information more directly accessible to the reader, we have revised the manuscript accordingly. The vertical profiles shown in Fig. 5 now display also the scaled number concentration of each particle type.

Figure 5c, d changed:

- *N_0·PF vertical profile added on top x-axis.*
- *PF vertical profile on bottom x-axis.*
- *Transparent dashed lines for N_0 · PF and solid line for PF.*

Figure 5 capture*: "[...] The black solid lines indicate isolines of PV (derived from ERA5) in PVU. The panel on the right shows the vertical profiles of PF for NO$^+$-rich (c), potassium-dominated particles (d) are shown for each phase as a function of potential temperature, independent from equivalent latitude. The bottom x-axis shows the PF with a solid line for each phase. The top x-axis shows the N0 ・PF of the particle types with a transparent dashed line for each phase. Details on the uncertainties are provided in Supplementary Information Sect. S3.5."*

L146: *"[...] The particle fraction (PF) for each particle type was calculated by binning the number of particles of each type relative to the total number of particles within 5K potential temperature and 5° equivalent latitude intervals. To additionally account for variations in the absolute abundance of ERICA-LAMS detected particles, particle type resolved number concentrations were calculated following Appel et al. (2022). Here, N_0 denotes the averaged ERICA-LAMS particle number concentration at the first detection stage in each bin. The scaled number concentration (N_0 ・PF) of a given particle type was obtained by multiplying N_0 with the corresponding PF. This quantity, N_0 ・PF, was used to evaluate whether changes in the relative contribution of a particle type are also reflected in changes in its number concentration within the ERICA-LAMS detected particle population. Comprehensive details regarding the ERICA-LAMS data post-processing and analysis are provided in the Supplementary Information Sect. S3."*

L263: *"[...] The increase in the PF of NO$^+$-rich particles during the Late Phase is mirrored by higher N_0· PF (Fig. 5c). Thus, the observed enhancement is not only reflected in the relative particle fraction, but also in the particle number concentration.[...]"*

L345: *"[...] The PF and N_0 ・PF of potassium-dominated particles decreased progressively from August (Early Phase) to September (Late Phase) (Fig. 5 b, d). [...]"*

Supplement after L125 (based on Appel et al., 2022):

(2) Scaled number concentration with N_0 · PF

The scaled number concentration was calculated by multiplying the particle fraction of a specific particle type PF with the averaged ERICA-LAMS particle number concentration at the first detection stage (N_0). The uncertainty was calculated *following the method described in Appel et al. (2022) and is presented here. The absolute uncertainty of* N_0 *was calculated using Poisson statistics:*

$$\sigma_{N_0}^{abs} = N_0 \cdot \sigma_{N_0}^{rel},$$

with the relative uncertainty given by:

$$\sigma_{N_0}^{rel} = \frac{1}{\sqrt{C_0}},$$

where C_0 *is the averaged ERICA-LAMS particle count at the first detection stage in the respective bin.*

The absolute uncertainty of the scaled number concentration was then calculated by Gaussian propagation of the uncertainties of PF and N_0*:*

$$\sigma_{PF \cdot N_0}^{abs} = \sqrt{(PF \cdot \sigma_{N_0}^{abs})^2 + (N_0 \cdot \sigma_{PF}^{abs})^2}.$$

Here, σ_{PF}^{abs} *is the absolute uncertainty of the particle fraction calculated from binomial statistics as described in Sect. S3.5(1), and* $\sigma_{N_0}^{abs}$ *is the absolute uncertainty of the averaged ERICA-LAMS particle number concentration at the first detection stage.*

3. **The description of the backward trajectories should be in a precise language (see also specific comments below).**

   See following response to comments below: 10, 11, and 13.

4. **The discussion of the influence of biomass burning and the measurements of organic aerosols are interesting but are in my opinion a bit away from the focus of this paper. The authors could reconsider if they really want to keep this part in the manuscript or if they want to present these findings in a different framework (it is just a suggestion from my side, both options would be okay for me).**

   Thank you for this suggestion. We agree that biomass burning is not the main focus of the manuscript. However, we decided to keep this discussion as it is, because it provides the most plausible explanation for the higher organic aerosol concentrations observed during the Early Phase. This interpretation is supported by the concurrent occurrence of potassium-dominated particles, which are commonly associated with biomass burning particles.

5. **Section 3.2 seems to be apart from the earlier part of the manuscript and introduces a new selection of data (also including more PHILEAS flights than mentioned in section 2.1, where only 6 flights were introduced, now also other flight data is used). Further, it is not shown how it is possible to compare air masses from 2017 with those measured in 2023. Is it known that those years are comparable? Are the air masses sampled during these campaigns comparable? Again, I suggest to consider if this section could be presented in a different framework than this manuscript. Further, the relationship if this section to the preprint by Koellner et al. (2026) is not very clear to me for this section.**

   We thank the reviewer for this important comment and for the critical assessment of Sect. 3.2. We agree that the original presentation could give the impression of a direct comparison between the StratoClim 2017 and PHILEAS 2023 measurements. Such a comparison is only possible to a limited extent because of the interannual variability of ASM dynamics, differences in the meteorological conditions, and differences in the sampled air masses. Our original intention was not to establish a direct year to year comparison, but to use the StratoClim 2017 measurements only as a chemical reference for the NO^+-rich particle type observed in the center of the

ASM (though within the ATAL). This seemed useful because these measurements were performed with the same instrument and characterized particles closer to their source region. However, we recognize that the way this was presented was potentially misleading and distracted from the main focus of the study.

To address this point, we revised Sect. 3.2. The direct comparison between StratoClim 2017 and PHILEAS 2023 was removed from the main figure and from the central discussion. The revised section now focuses on the evolution of the internal mixing state of NO^+-rich particles within the PHILEAS measurements. In particular, the discussion now follows the transition from particles observed in AMA filaments toward particles mixed into ExLS background air masses. This revision links the section more directly to the main objective of the manuscript, namely the influence of ASM outflow on the aerosol composition of the ExLS during late summer and autumn 2023.

We also clarified the data selection used in this section. The additional PHILEAS flights are now explicitly described as part of a targeted analysis of NO^+-rich particle composition and internal mixing state, rather than as part of the Early and Late Phase comparison introduced in Sect. 2.1. This distinction has been clarified in the revised text and in the figure description.

Furthermore, we clarified the role of the preprint by Köllner et al. (2026). In the revised manuscript, this study is not used as a direct basis for comparison with our data, but as independent supporting evidence that pollution aerosol, including ammonium nitrate, can be transported northward from the ASM region and irreversibly mixed into the ExLS. We therefore refer to Köllner et al. (2026) only in this context, namely to support the interpretation that the observed NO^+-rich particles in PHILEAS are consistent with ASM related aerosol transport into the ExLS.

To retain the useful chemical reference provided by the StratoClim 2017 measurements without distracting from the main argument of the manuscript, we moved the modified version of the former Fig. 10 to the supplement. The corresponding discussion in the main text was shortened and revised accordingly. The supplement figure now serves only as supplementary chemical context for the NO^+-rich particle type and is no longer presented as evidence for a direct comparison between 2017 and 2023 air masses.

L137: *"[...] The particle size selection is based on the same particle types defined in Appel et al. (2022) (StratoClim campaign in 2017), which also serves as the basis for particle selection in this study.[...]"*

L357-L360: *„First,* ~~*we analyzed the NO^+-rich particles measured within the center of the AMA during the StratoClim 2017 mission in order to establish a reference for this particle type in close proximity to its source. Particularly, we analyzed the data below the 370 K potential temperature (defined as 'lower ATAL' in Appel et al., 2022), to study the NO^+-rich particles not affected by stratospheric air masses.*~~

L372: *These* three *phases represent different extents of chemical processing during atmospheric transport and residence time, providing a framework for interpreting the evolution and mixing state of NO^+-rich particles* ~~*from the center of the AMA into the ExLS*~~.

L378: *“[…] First, the comparison between the AMA filament and the Late Phase shows a broadly similar pattern in the selected m/z signal ratios (Fig. 11). All three ratios are slightly higher in the Late Phase than in the AMA filament, indicating modest compositional changes during transport from the AMA region into the ExLS. However, the overall structure of the ratios remains largely preserved. This suggests that the transport of AMA filament air masses into the ExLS and its subsequent mixing with the stratospheric background did not yet lead to strong chemical processing of these NO^+-rich particles. Second, a significant increase in the ammonium-to-nitrate ratio and decrease of the nitrate-to-sulfate ratio from the Late to the Early Phase points to changes in the internal mixing state of NO^+-rich particles, with a shift toward lower nitrate and/or higher ammonium and sulfate content. This change in the internal mixing state of NO^+-rich particles likely results from continued inclusion of sulfuric acid through condensation of gaseous sulfuric acid and/or coagulation with sulfate-rich particles, together with the depletion or replacement of particulate nitrate through chemical processing (e.g., Kremser et al., 2016; Junge and Manson, 1961; Murphy et al., 2014; Schneider et al., 2021).[…]“*

L379-L398: *Moved to the supplement and updated to match the new figure.*

L416: *„[…] Furthermore, we analyzed the internal mixing state of NO^+-rich particles measured in the center of the ASM region (within the ATAL) during the StratoClim 2017 campaign (see Supplement Information Sect. S7). These results showed even less chemically processed aerosol during the StratoClim 2017 campaign in the center of the ATAL, where particles were freshly formed below 380 K in the upper troposphere (see Fig. S9). Since interannual variability between 2017 and 2023 may play a significant role, the observation should be interpreted with caution. However, it does not rule out the hypothesis that a chemical aging from the center of the ATAL to the ExLS is possible with longer atmospheric residence time.”*

We also revised the description of the additional PHILEAS flights F08 to F18 used in this section. These data are not used for the Early and Late Phase comparison of ExLS background air masses introduced in Sect. 2.1. Instead, they are used specifically to identify AMA filament air masses within the ExLS. The selection is based on enhanced CH_4 mixing ratios as an indicator of Asian boundary layer influence, together with an additional N_2O criterion to restrict the analysis to stratospheric air masses. This clarification now more clearly explains why a different PHILEAS flight selection was used for this part of the analysis.

We retained the definition of AMA filament air masses given in lines 360 to 366, including the CH_4 and N_2O thresholds. This definition is now explicitly linked to the purpose of this section, namely to examine the chemical characteristics and internal mixing state of NO^+-rich particles in air masses influenced by the ASM outflow, rather than to extend the Early and Late Phase comparison.

L106: *“[…] For the subsequent analysis of particle composition, the AMA filaments measured during the PHILEAS campaign from flights F08 to F18 in the ExLS were included. These flights were specifically selected because they represent transport from the AMA region and thus also the influence on the extratropics, as shown and discussed in the studies by Köllner et al. (2026) and Riese et al. (2025). The flight paths of these selected flights are shown in the Supplementary Information Fig. S1.[…]“*

In addition, we clarified the role of Köllner et al. (2026) in the revised manuscript. This accompanying study investigates AMA filaments during PHILEAS with a stronger focus on transport processes and model analyses. Our study therefore does not use Köllner et al. (2026) as a prerequisite for the analysis presented here, but rather as independent supporting evidence for the interpretation of the PHILEAS in-situ aerosol measurements. The model analyses presented by Köllner et al. (2026) are consistent with our interpretation that particles containing ammonium nitrate undergo chemical processing during transport and residence in the ExLS. In particular, they support the interpretation of a relative decrease in nitrate and an increasing importance of sulfate as the particles become more internally mixed during transport from AMA filament air masses into the ExLS background.

L443: *„[…] The results of our in-situ measurements are consistent with the results of the study by Köllner et al. (2026), describing the transformation of ammonium nitrate to ammonium(bi) sulfate.[…]“*

Specific points:

6. **very minor point: The numbering in the list of affiliations is not according to the appearance in the authors list. At the moment, numbers are counting: 1,2,5,3,10,8,7,9,4,6**

   Done.

7. **L113: Please define HASI**

   We thank the reviewer for this comment. We have now rephrased and defined HASI explicitly as the HALO Aerosol Submicrometer Inlet and added a short description of its function as a forward-facing aerosol inlet used for sampling submicron aerosol particles aboard HALO.

   L111: *"[...] Aerosol particles outside the aircraft were sampled using the HALO Aerosol Submicrometer Inlet (HASI), which is a forward-facing aerosol inlet. This inlet system was developed specially for sampling submicron aerosol particles aboard the HALO aircraft (Minikin et al., 2017; Andreae et al., 2018) and was combined with the HASI Flow Control Unit (FCU).[...]"*

8. **L233: Why is only a degraded ERA5 resolution used for the PV analysis?**

   The PV analysis shown in this study is based on ERA5 data with a horizontal resolution of 1° × 1°. We would like to clarify that the PV values are not used as the primary criterion for the main scientific conclusions. The identification of stratospheric background air is based on the in-situ trace gas measurements, using the N_2O and CO thresholds described in Sect. 2.3.2. The ERA5 derived PV values and equivalent latitudes are used only as an additional consistency check for these thresholds and as contour lines in the figures to support the dynamical interpretation.

   Furthermore, during the initial analysis steps, the CLaMS data were calculated using ERA5 meteorological data with a horizontal resolution of 1° × 1°. Later on, the CLaMS data were recalculated using the higher-resolution ERA5 data with 0.3° × 0.3°. The CLaMS data were specially calculated and provided upon request, which is why we received the final version with a higher resolution (0.3° × 0.3°). Therefore, the main conclusions regarding transport pathways and source regions are not based on the PV field with a resolution of 1° × 1°, as was incorrectly mentioned in lines L232–L234.

   The ERA5 data provided for the campaign, with a slightly lower resolution (1° × 1°), are publicly available on Zenodo (https://doi.org/10.5281/zenodo.15076519). They were used for the calculation of PV and equivalent latitude. In our opinion they are sufficient for a comparison between the tracer measurements and the dynamically modeled boundary condition. To avoid confusion, we will clarify this point in the manuscript:

   L232-L234: *"[...] The PV values and equivalent latitudes were derived from the ERA5 data with 1°x1° horizontal resolution. The PV values consistently exceed 7 PVU with major contribution from PV above 8 PVU when N_2O and CO concentrations are below 336 ppbv and 40 ppbv, respectively. [...]"*

__L235:__ "These PV values are used only as an additional consistency check for the N_2O and CO based classification and for the dynamical interpretation of the sampled air masses. The transport and origin analysis itself is based on CLaMS data driven by ERA5 meteorological data with a horizontal resolution of 0.3°x0.3°.

__L366:__ "[...] Since the focus was the transport of the ASM-influenced air masses into the ExLS, the selection of AMA filaments has been restricted to the stratospheric region with these trace gas thresholds. Therefore, no further threshold, such as potential temperature, was used as a criterion for this selection. [...]"

9. **Figure 4 d-f: Are the profiles shown in the right column the sum or an average of all equivalent latitudes? Same for Figure 5.**

Thank you for pointing this out. For Fig. 4d to f, the profiles show the mean ERICA-AMS mass concentration for each potential temperature bin and for each campaign phase separately. All data points fulfilling the applied ExLS background air mass criteria (N_2O < 336 ppbv and CO <40 ppbv) were assigned to the respective potential temperature bin, irrespective of their equivalent latitude. The mean concentration was then calculated from all individual measurements within that potential temperature bin. Thus, the profiles are not calculated as arithmetic averages over equivalent latitude bins, but represent vertical mean concentrations based on all sampled data points within each potential temperature bin.

For Fig. 5c and d, the same binning principle was applied to the ERICA-LAMS data. However, the profiles show particle fractions rather than mean mass concentrations. For each potential temperature bin and campaign phase, all detected particles were binned over the sampled equivalent latitude range. The particle fraction was then calculated as the number of particles of a given type divided by the total number of detected particles in the same potential temperature bin. Thus, the Fig. 5 profiles are count based particle fractions as a function of potential temperature, not averages over equivalent latitude bins.

__In caption Fig. 4:__ "[...] Panels (d–f) show the corresponding average mass concentrations of nitrate (d), ammonium (e), and organic matter (f) for the Early (blue) and Late (orange) Phases as a function of potential temperature, independent from the equivalent latitude. [...]"

__In caption Fig. 5:__ "[...] The black solid lines indicate isolines of PV (derived from ERA5) in PVU. The panel on the right shows the vertical profiles of PF for NO^+-rich (c), potassium-dominated particles (d) are shown for each phase as a function of potential temperature, independent from the equivalent latitude. [...]"

10. **Figure 8: I do not understand the "relative frequency" shown in this figure. Relative with respect to what? And are the values normalized with respect to a surface area? And what is meant by "backward trajectory end point"? The point where the trajectory ends (at the position of the aircraft), or the point where the backward trajectory ends (at the surface)? And the "relative frequency" shown in the figures - is it of backward trajectories ending at the lowest model level or what criterion was used here that a trajectory is shown? Further, 0.03% of all trajectories is a very, very low fraction of trajectories being the top of the color bar. I cannot integrate over the area to get the total fraction of trajectories which were (probably?) reaching the ground, but it seems like most of the backward trajectories did not reach the ground during the maximum time period?**

    For clarifying some confusion, the following text describes the handling with CLaMS backward trajectories for this plot:

    For each phase, the trajectories were initialized along the HALO flight path for air masses classified as ExLS background air using the criteria $N_2O < 336$ ppbv and CO < 40 ppbv. The backward trajectories until 1 May 2023 were used. For Fig. 8, all trajectory positions with potential temperatures between 320 K and 380 K were selected and counted in 1° × 1° longitude and latitude grid boxes. The relative frequency in each grid box is defined as the number of counted trajectory points in each grid box divided by the total number of trajectory points within the potential temperature interval.

    We agree that the terminology and the normalization of Fig. 8 were not sufficiently clear. The figure does not show backward trajectory end points, and it does not show only trajectories reaching the surface or the lowest model level. Instead, it shows all positions along the CLaMS backward trajectories that fulfill the selected potential temperature criterion (e.g., all trajectory points between 320-380K). In this context, we used the wrong term for "endpoints" and have corrected the whole section in the manuscript.

    The counted trajectory points within the grid boxes are normalized to the total number of trajectory points within the potential temperature range from 320 K to 380K and are shown in the respective panel. They are not normalized with respect to the physical surface area of the grid boxes; instead, they represent the point density per angular area (1°x1°).

    The maximum value of ~0.03% on the color bar may appear low, but it represents the fraction within a single 1°x1° bin. Summing these values over the observed high-density regions (e.g., the Asian Monsoon Anticyclone core) accounts for a significant portion of the total air mass. The figure is intended to show the spatial residence time of the backward trajectories within the ExLS potential temperature buffer, rather than a 1D fraction of trajectories reaching the boundary layer.

    However, we used this comment as an opportunity to simplify both the trajectory analysis shown in the main text and the corresponding discussion:

    In the previous version of the manuscript, the trajectory density plots shown in the main text were based on trajectory points within selected potential temperature ranges. These plots were intended to illustrate the transport pathways of the sampled air masses within the UTLS and to provide a dynamical context for the vertical profiles of ERICA-AMS and ERICA-LAMS. This perspective is also consistent with the vertical profile analysis by Appel et al. (2022). Furthermore, this is also consistent with Köllner et al. (2026), who point out that quasi-horizontal transport within these potential temperature ranges is important for the redistribution of air masses influenced by the ASM.

However, we agree that these plots may distract from the main question addressed in this part of the manuscript, namely the lower tropospheric origin of the sampled air masses. Therefore, we moved the trajectory plots based on potential temperature ranges (350-380 K corresponding to quasi-horizontal transport) from the main text to the Supplementary Information. They are retained there as supporting information because they remain useful for comparing the transport pathways with the potential temperature-based aerosol profiles.

In the revised main text, we now show a simplified trajectory analysis that focuses on trajectories reaching the boundary layer. This is done by ending the trajectories a vertical hybrid coordinate (ζ) = 120 K. The model parameter, ζ is used for the CLaMS simulations, which follows potential temperature in the stratosphere and gradually transforms into a pressure based coordinate in the troposphere, there by accounting for surface pressure and orography (Pommrich et al., 2014). Following Vogel et al. (2026), ζ = 120 K was used as threshold for the model boundary, which corresponds to an altitude of approximately 2 to 3 km above the Earth's surface. All trajectories at ζ = 120 K are counted in 1° × 1° latitude longitude bins. This representation provides a clearer indication of the lower tropospheric regions connected to the sampled Early and Late Phase air masses. The revised figure therefore better supports the discussion of air mass origin, while the Supplementary Information provides the additional UTLS transport perspective. Both plots omit the relative frequency of the trajectory points and instead show the number of trajectory points within each grid box to provide a clearer overview.

L212: *"[...] However, the initial position of the CLaMS backward trajectories along the aircraft flight path were selected separately for the Early and Late Phases, based on the trace gas criteria for ExLS background air masses defined in Sect. 2.3.2. The trajectories were then traced backward until they reached a vertical hybrid coordinate (* ζ *) = 120 K. Here,* ζ *is used for the CLaMS simulations, which follows potential temperature in the stratosphere and gradually transforms into a pressure based coordinate in the troposphere, there by accounting for surface pressure and orography (Pommrich et al., 2014). Following Vogel et al. (2026),* ζ *= 120 K was used as threshold for the model boundary, which corresponds to an altitude of approximately 2 to 3 km above the Earth's surface. Finally, all backward trajectories reaching this threshold were defined as trajectory endpoints and counted within 1°x1° latitude and longitude grids. This approach was used to identify low tropospheric source regions that were connected to the studied ExLS background air masses via backward transport.[...]"*

L293-L308: *"[...] In addition, Fig. 9 shows CLaMS-derived backward trajectory statistics for air masses observed during the Early Phase (Fig. 9a) and Late Phase (Fig. 9b). It is demonstrated that differences in backward trajectory endpoints (trajectories reached the boundary layer) exists between the Early and Late Phases, indicating changes in the predominant source regions and transport characteristics of young air masses reaching the ExLS. The Late Phase, in comparison to the Early*

Phase, exhibits a pronounced, spatially coherent maximum in trajectory endpoints over northern Indian Subcontinent and along the southern margin of the Tibetan Plateau. It is thus obvious that the ExLS background air during the Late Phase was strongly influenced by relatively young (< 3.5 months) air masses from ground-level sources in South-East Asia. Additional trajectory analyses, shown in the Supplementary Information Sect. S6, indicate that within the potential temperature range of 350 to 380 K, the trajectories suggest the AMA acts as a reservoir for ASM-influenced air. This allows the masses to reside for extended periods before being exported poleward. The enhanced nitrate concentrations observed during the Late Phase should therefore not be interpreted solely as a local accumulation of particulate nitrate with time. Instead, they likely reflect the progressive seasonal accumulation of ASM-influenced air masses in the ExLS, driven by repeated eddy shedding and air mass transport from the AMA, which is most frequent during July and August but continues into September (e.g., Clemens et al., 2022; Vogel et al., 2016, 2019). This result goes along with a recent study by Köllner et al. (2026), which provides direct evidence for the northward transport of pollution aerosol (including ammonium nitrate) from the ASM region and irreversible mixing of this aerosol into the ExLS. The authors conclude that the quasi-horizontal isentropic advection occurring above 370 K potential temperature in the lower stratosphere is the most important transport pathway of pollution aerosol associated with the ASM region into the ExLS. In combination with their findings, this study provides additional evidence that polluted aerosol from Asia containing ammonium and nitrate are enriched in the ExLS background air and can persist there for a few months.[…]"

<u>Supplement L224-L241:</u> *"[…] S6. CLaMS Backwards Trajectories for Early and Late Phase*

The CLaMS backward trajectories were initialized along the flight track of each PHILEAS flight. All back trajectories are separated into two groups according to the PHILEAS Early and Late Phases, as described in detail in the main text. The trajectories shown in Fig. S8 were selected based on the potential temperature range of 350 to 380 K, and the trajectory counts were not normalized by the physical grid box area. The maps therefore show the number points of trajectory positions in regular 1°x1° longitude and latitude grid boxes, rather than an area weighted density. This choice was made because the analysis focuses on identifying preferred transport pathways and regions of enhanced trajectory occurrence. Since the main trajectory locations discussed here are located mainly in tropical and subtropical regions, the variation in physical grid box area due to the convergence of longitudes with latitude is comparatively small and does not affect the qualitative interpretation of the transport patterns. The trajectories shown in Fig. S8 are restricted to the potential temperature range from 350 to 380 K. This range represents the lower stratospheric transport regime in which quasi-horizontal transport from the AMA into the ExLS is expected to occur."

Figure S8: "CLaMS backward trajectories located between 350 K and 380 K potential temperature during the Early (a) and Late Phases (b) of the PHILEAS campaign. The value in each grid box represents the number of selected trajectory positions in that grid box. The values are not normalized by grid cell area. Thus, the figure shows the horizontal occurrence of trajectory positions within this potential temperature range *and does not indicate the fraction of trajectories reaching the surface or the lowest model level.”*

11. **309: What is meant by: "backward trajectory statistics were also examined for potential temperatures below 320 K and above 380 K"? Backward trajectories being released at measurement locations of this potential temperature range? I think the formulation of the description of the trajectories could be more precise throughout the manuscript.**

    It means all the backward trajectory points below 320K potential temperature from measurement location back until 1 May 2023 for each Phase (Early or Late Phase). This plot was anyhow removed from the manuscript as it provides no additional information. Instead, it was replaced by (now) Fig. 9, showing trajectories reaching the boundary layer (see comment 10).

12. **L323: I would argue that the peak of CO PDF is only marginally shifted towards higher mixing ratios, while also lower mixing ratios appear in the PDF. "The CO distributions (Fig. 9b) show a similar pattern." is not quite precise here.**

    Thank you for this comment. We agree that the wording was too strong and that the CO distribution does not show the same clear shift as CH_4. We have therefore revised the sentence to state that the Late Phase CO distribution is only marginally shifted toward higher mixing ratios, while both phases strongly overlap and lower CO mixing ratios are also present during the Late Phase.

    L323: *“[...] Compared to CH_4, the difference between the Early and Late Phase CO distributions is less pronounced (Fig. 10b). The Late Phase CO distribution is only marginally* shifted toward *higher mixing ratios, while both distributions strongly overlap and lower CO mixing ratios are also present during the Late Phase. Overall, the distribution appears broader in the Late Phase than in the Early Phase and a local maximum of the probability density in the Late Phase occurs near 30–32 ppbv, whereas the Early Phase peaks closer to 27–29 ppbv. [...]”*

13. **L347: "This trend suggests a decreasing influence of BB emissions on the ExLS aerosol composition": Is this also supported by the trajectories shown in Fig. 8? For me it looks like there is more signal from Canada in the early phase compared to the late phase.**

    We thank the reviewer for this important comment. We agree that the previous wording may have been too strong and that the trajectory analysis shown in the former Fig. 8 could give the impression of an enhanced connection to Canada during the Early Phase. We therefore revisited this interpretation and clarified the description of the trajectory analysis in the revised manuscript (see comments before).

    In the previous version, the figure caption and the corresponding text did not sufficiently clarify that the displayed trajectory density does not represent boundary layer source regions or trajectory endpoints at the surface. Instead, it shows backward trajectories within a defined potential temperature range. Therefore, enhanced trajectory density over Canada in this figure indicates that air masses passed through this region within the selected potential temperature range, but it does not necessarily imply direct uptake of biomass burning emissions from the Canadian boundary layer.

To make this distinction clearer, we revised the text and figure description (see comment 10). In addition, the new Fig. 9 now shows the trajectory analysis based on boundary layer contact. This analysis indicates a clear contribution from the ASM source region, while no comparable boundary layer contribution from Canada or the broader extratropical region is observed for the selected air masses. This supports our interpretation that the enhanced nitrate signal is primarily linked to ASM-influenced air masses rather than to direct boundary layer uptake from Canadian biomass burning sources.

14. **L360: "Second, we analyzed NO^+-rich particles abundant in CH_4-rich stratospheric regimes in the extratropics measured during PHILEAS (defined as 'AMA filament in the ExLS').": Is there also a potential temperature threshold applied to this selection of air masses?**

    We thank the reviewer for pointing this out. No explicit potential temperature threshold was applied to this selection. Instead, the "AMA filament" air masses were identified based on enhanced CH_4 mixing ratios, using CH_4 > 1920 ppbv following Köllner et al. (2026) and Tomsche et al. (2019). Since our study focuses on transport into the ExLS, we further restricted the selection to stratospheric air masses by applying an additional N_2O criterion of $N_2O < 336$ ppbv. This approach allows the selection of stratospheric air masses with a clear Asian boundary layer influence without imposing an additional dynamical altitude constraint. We added a short explanation to clarify this in the revised manuscript.

    L365: *We added a reference to the new supplement figure (Fig. S1) showing the flight path overview (F08–F18). L366: (focus on the western Pacific, northern Canada and Alaska region - Köllner et al., 2026, Fig. S1)*

    L365: *"[…] Since the focus was the transport of the ASM-influenced air masses into the ExLS, the selection of AMA filaments has been restricted to the stratospheric region with these trace gas thresholds. Therefore, no further threshold, such as potential temperature, was used as a criterion for this selection. […]"*

    Supplement addition L1: *Figure S1 provides an overview of the flight tracks conducted during the PHILEAS campaign in 2023. The figure complements the description of the air mass selection used for the analysis of AMA filament air masses in the ExLS. In addition to the six flights used for the Early and Late Phase comparison in the main text, the filament analysis also includes measurements from flights F08 to F18. These flights sampled air masses over the western Pacific, Alaska, northern Canada, and northern Europe, where AMA filament were identified based on enhanced CH_4 mixing ratios following Köllner et al. (2026). The flight path overview is therefore included to provide spatial context for the broader flight selection used in this part of the analysis.*

15. **L407: Make a new paragraph at "Third"**

    Done.

16. **L409: "The size distribution of the Early Phase peaks at larger diameters": I would argue that all three distributions peak at the same particle diameter (of ~340 nm) in Fig. 11!**

    Thank you for pointing this out. We agree that all three distributions show their main maximum at approximately 340 nm. The original wording was therefore misleading. We will revise the sentence and state instead that the Early Phase shows a broader size distribution with a stronger relative contribution from larger particle diameters compared to the AMA filament and Late Phase.

    L409: *"Third, the ERICA-LAMS-derived particle size distribution of NO^+-rich particles (Fig. 12) shows a noticeable change toward larger particle diameters in the Early Phase compared to both the Late Phase and the AMA filament. The size distribution of the Early Phase shows a broader distribution and a stronger relative contribution of larger particle diameters compared to the AMA filament and the Late Phase measurements. This broader distribution toward larger particle diameters in the Early Phase is consistent with progressive particle growth during longer residence times in the stratosphere. Further it supports the conclusion that the NO^+-rich particles present during the Early Phase have undergone more chemical and microphysical aging in the stratospheric background. Taken together, the ERICA-LAMS ion signal ratios and particle size as well as CLaMS model data provide compelling evidence that the Early Phase of PHILEAS was dominated by chemically aged NO^+-rich particles within the air masses of the ExLS background. […]"*

17. **Figure 10: This is a busy plot and I think it should be mentioned why the "Early Phase" has a grey background, and why there are no green bars for the "Filament" and "Late" columns, and why there is no blue bar for the "Early Phase".**

    We thank the reviewer for this comment. We agree that the original Fig. 10 was too crowded and that the graphical elements were not sufficiently explained. In particular, the grey background for the Early Phase and the absence of some bars could lead to confusion.

    After further discussion and careful reevaluation of the figure, we decided to revise this analysis and the corresponding figure substantially. In the previous version, the threshold of 5 mV·samples, which was introduced in the methods section and in the supplement to identify noisy spectra, was also applied to individual ion signals used for the calculation of signal intensity ratios. This was not appropriate for this specific analysis. The threshold was originally defined to exclude spectra whose highest peak did not exceed 5 mV·samples. Its purpose was therefore to remove spectra dominated by noise, not to filter individual ion signals within otherwise valid spectra.

    Applying this threshold to each individual signal before calculating the ratios artificially affected the resulting ratios and biased the comparison between the different air mass categories. This also explains why some bars were absent in the original figure. We therefore recalculated the signal intensity ratios without applying the 5 mV·samples threshold to individual ion signals. This provides a more representative picture of the relative signal contributions within the valid NO^+-rich particle spectra.

    In addition, and in response to the more general concern raised in comment 5, we reconsidered the direct comparison between StratoClim 2017 and PHILEAS 2023. We agree that such a comparison is limited by interannual variability, different meteorological conditions, and differences in the sampled air masses. Therefore, we removed the direct StratoClim 2017 comparison from the main figure and from the central discussion in the main text. The revised main figure now focuses only on PHILEAS 2023 and on the development of the NO^+-rich particle composition from AMA filament air masses toward ExLS background air masses.

The modified StratoClim comparison was moved to the Supplementary Information, where it is now presented only as additional chemical context for the NO^+-rich particle type observed with the same instrument. The revised text explicitly states that this comparison should be interpreted with caution as a direct comparison between the sampled air masses in 2017 and 2023.

New Caption of Figure 10 (now Figure 11): *"Ratios of key ion peaks in NO^+-rich particles across different PHILEAS phases. The green bar represents the ratio of less oxidized to more oxidized organic components, highlighting changes in organic composition over time. The orange bar represents the ammonium-to-nitrate signal intensity ratio. And the blue bar represents the nitrate-to-sulfate signal intensity ratio. The arrow at the bottom indicates the temporal evolution and chemical processing of the NO^+-rich particles within the AMA filaments/Late Phase to the Early Phase, emphasizing how long-term stratospheric residence alters particle composition. The gray dashed area in the Early Phase is a visual marker to illustrate that the Early Phase differs from the other phases in terms of its long-term residence time in the stratosphere and the associated impact on the single particle composition. More information about the uncertainty in Supplement Information Sect. S3.5."*

18. **L446: "We thus hypothesize that the particles may remnants from the previous monsoon season.": Please rephrase, I do not understand this sentence.**

We thank the reviewer for pointing this out. We agree that the original sentence was unclear and grammatically incorrect. We have rephrased it to clarify that we refer to chemically aged aerosol remnants that may have persisted in the ExLS from the previous monsoon seasons until the Early Phase of PHILEAS.

L446: *"[...] We hypothesize that the aged aerosol particles measured during the Early Phase of the PHILEAS campaign originated from the previous ASM seasons and may have remained in the stratosphere for months to years. [...]"*

19. **L450 & Fig. 12: It is not appropriate to introduce new figures in the section called "conclusions". Please introduce this figure in the main part of the manuscript.**

We thank the reviewer for this comment. We agree that the schematic figure should not be introduced for the first time in the conclusions. We have therefore moved the figure to the main results and discussion section, specifically to the subsection discussing the ASM outflow as the driver of the enhanced nitrate and ammonium aerosol in the ExLS. The figure now provides a conceptual framework for the subsequent interpretation of the ERICA aerosol measurements and the CLaMS model results.

L272: *"[...] Figure 6 provides a schematic overview of the large-scale AMA circulation and the associated filamentary transport pathway toward the ExLS. [...]"*

__Figure 5 caption (now Figure 6):__ *"The illustration highlights the large-scale circulation of the AMA and its transport pathway toward the ExLS. It is based on ERA5-derived geopotential height at 100 hPa averaged over July and August 2023. The orange contour marks the region where ozone mixing ratios reach approximately 200 ppbv, indicating the transition toward the ExLS. Schematic arrows highlight the anticyclonic flow within the ASM, as well as the transport of the AMA filament that carries ASM-influenced air masses towards higher latitudes, subsequently impacting the ExLS region. The cycle arrows demonstrate the filament mixing into the ExLS background."*

__L449-L450:__ *"[...] Altogether, our study confirms the critical role of the ASM circulation as a significant pathway for transporting aerosols into the extratropical region, subsequently affecting the ExLS aerosol composition ~~(as illustrated schematically in Fig. 12)~~ [...]"*

20. **L454: Here, year-to-year variability is mentioned, but this manuscript directly compares measurements from 2017 to those from 2023. Why is that a valid comparison?**

We thank the reviewer for pointing this out. As already addressed in our responses to comments 5 and 17, we agree that a direct comparison between StratoClim 2017 and PHILEAS 2023 is limited by interannual variability in ASM dynamics, differences in meteorological conditions, and differences in the sampled air masses.

To avoid this ambiguity, we removed the direct comparison between the 2017 and 2023 measurements from the main text. The revised manuscript no longer uses the StratoClim 2017 data as direct evidence for the interpretation of the PHILEAS 2023 observations. Instead, the PHILEAS based discussion now focuses on the development of the NO^+-rich particle type within the PHILEAS 2023 measurements.

The former comparison with StratoClim 2017 was moved to the Supplementary Information, where it is now presented only as additional chemical context for the NO^+-rich particle type observed with the same instrument. We also revised the corresponding text to make clear that this comparison is not used to draw conclusions about interannual differences or about the comparability of the sampled air masses in 2017 and 2023.

__Supplement L242:__ *"*

S7. Chemical Composition Changes of NO^+-rich Particles from the ATAL to the ExLS

The NO^+-rich particles measured within the AMA during the StratoClim 2017 mission were analyzed to provide a compositional reference for this particle type closer to its source region. In particular, we selected particles measured below 370 K potential temperature, corresponding to the lower ATAL as defined by Appel et al. (2022). This selection was used to characterize NO^+-rich particles in the upper tropospheric part of the ATAL, where the influence of older stratospheric background air is expected to be smaller.

Figure S10 summarizes the selected ion signal ratios for NO^+-rich particles in the lower ATAL from StratoClim and compares them with the PHILEAS phases discussed in the main text. The comparison among the AMA filament, the Late Phase, and the Early Phase is described in detail in Sect. 3. Here, the StratoClim lower ATAL data are used as an additional reference to place the PHILEAS *observations into a broader chemical context. Compared with the lower ATAL reference, the AMA filament observed during PHILEAS shows a much lower nitrate-to-sulfate ratio and a clearly lower ratio of less-to-more oxidized organic fragments. This indicates that NO^+-rich particles in the AMA filament air masses sampled in the ExLS were already chemically processed relative to particles observed within the lower ATAL. In contrast, the ammonium-to-nitrate ratio remains similar between the lower ATAL and the AMA filament, suggesting that the most pronounced differences are associated with the relative sulfate contribution and the oxidation state of the organic component. The Early Phase shows the strongest deviation from the lower ATAL reference. In particular, the ratio of less-to-more oxidized organic fragments is substantially lower, indicating a stronger contribution of more oxidized organic fragments. This supports the interpretation that NO^+-rich particles in the aged ExLS background (Early Phase) had undergone additional chemical processing during prolonged residence in the stratosphere. The very low nitrate-to-sulfate ratio in the Early Phase further suggests a stronger relative sulfate signature compared with both the lower ATAL and the more recently transported PHILEAS air masses.*

However, this comparison has to be interpreted with caution. The StratoClim and PHILEAS measurements were performed in different years and during different ASM seasons. Therefore, the lower ATAL particles measured in 2017 cannot be treated as a direct precursor of the AMA influenced particles observed during PHILEAS in 2023. Interannual variability in the strength, transport pathways, emission patterns, and chemical processing within the ASM may affect the comparability of both data sets. Consequently, the StratoClim data are used here only as a qualitative reference for NO^+-rich particles closer to the ATAL source region. Within this limitation, the observed differences support the interpretation that transport from the AMA into the ExLS and subsequent stratospheric residence are associated with increasing chemical aging, especially with respect to organic oxidation and the relative sulfate contribution. "

21. **L455: The authors recommend further campaigns to answer open questions, but are there options for long-term observations from operational monitoring programs like from satellites or regular aircraft missions?**

We agree that long-term observations from satellites and regular aircraft programs are valuable for investigating the seasonal and interannual variability of aerosol layers, transport pathways, and trace gas distributions in the UTLS. We will clarify this in the conclusion. However, for the specific chemical questions addressed in this study, targeted aircraft-based in-situ measurements remain essential. Satellite observations provide broad spatial and temporal coverage, but they do not resolve the detailed submicron aerosol composition and particle mixing state needed to distinguish ammonium nitrate, sulfate, organic matter, and biomass burning related particles in the ExLS. Regular aircraft programs can provide important long-term context, for example through trace gas and aerosol observations, but they typically do not provide the same level of single-particle chemical information as ERICA-LAMS. Therefore, future work should ideally combine long-term observational programs with dedicated in situ campaigns.

<u>L453:</u> *"Future progress would therefore benefit from combining long-term observational programs with dedicated aircraft based in-situ measurement campaigns. Satellite missions such as EarthCARE can provide a global perspective* *on aerosol layers and large-scale transport pathways in the UTLS, while regular aircraft programs such as IAGOS CARIBIC can provide valuable long-term in-situ information on trace gases and aerosol composition. However, targeted aircraft campaigns remain essential for resolving the detailed submicron aerosol composition, single particle mixing state, and chemical aging of the ASM-influenced aerosol particles in the ExLS. Therefore, future studies should combine satellite monitoring, regular aircraft observations, global chemical transport modeling, and continued airborne single particle mass spectrometry measurements. In addition, laboratory investigations are needed to better constrain the chemical aging mechanisms of NO^+-rich particles, their atmospheric evolution, and the timescales of the relevant chemical processes. […]"*

22. **L461: I would appreciate if the authors would make their data available to the referees. There are options to publish the data password protected during review if they prefer not to make their data publicly available during the review process. In my opinion, it is also part of the referee's job to assess the quality of the published data, which belongs to this paper, and I cannot do so at the moment.**
    - ERA5 Data: Uploaded on Zenodo (https://doi.org/10.5281/zenodo.15076520)
    - BCPD data include cloud flags (with "1" for cloud interaction and "0" for no clouds during measurements).
    - ERICA-AMS/-LAMS, UMAQS and CLaMS: will be uploaded on seafile (password and link will be handout to the editor)

General changes:

- The numbering of the Supplement sections was adjusted because a new Section S1, `Flight path overview of the PHILEAS campaign 2023', was added.
- A description of the data availability has been included in the revised manuscript.
- The citation of Vogel et al. (2026) was corrected, as the study has now been published.
- The order of the affiliations was corrected.
- Figure 12 in the original manuscript is now shown as Figure 6 in the revised manuscript. Consequently, the numbering of the subsequent figures was adjusted accordingly.

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