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 #1** (https://doi.org/10.5194/egusphere-2026-998-RC1) 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. **One important conclusion of this paper is regarding the transport of ATAL materials to ExLS that NO3 is higher in Sep than other time. I wander if you could add more insight in the paper regarding following questions, I am curious about: Is the peaking of NO3 in ExLS just because the cumulation of NO3 takes time? Or is it because in August, there're more shedding events that "leaks" more materials out of the ASM anticyclone compared to previous months? When ASM anticyclone breaks, more ATAL materials gets to transport to ExLS or ATAL materials has no chance to enter the stratosphere?**

   We thank the reviewer for this insightful comment. We agree that the enhanced nitrate observed in September should not be interpreted simply as a result of local nitrate accumulation with time. Rather, the increase reflects the progressive accumulation of ASM-influenced air masses in the ExLS during the monsoon season, caused by repeated eddy shedding events and filamentary export from the AMA. Climatological analyses show that AMA cutoff activity starts in June, peaks in July and August, and remains frequent in September, thereby enabling continuous quasi-horizontal transport of the ASM-influenced air masses into the ExLS (Clemens et al., 2022).

   Previous CLaMS studies have also demonstrated the seasonal buildup of air masses from the ASM region in the ExLS and showed that the breakup of the anticyclone in September supports the export and redistribution of AMA air into extratropical regions. This interpretation is consistent with our results, which indicate a stronger contribution of relatively young air masses from South Asia and the western Pacific during the Late Phase of PHILEAS. Therefore, the higher nitrate concentrations in September are most likely linked to the cumulative export of aerosol associated with the ASM region. We have further clarified this point in the revised the manuscript as follows:

   L301: *"[...] 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). [...]"*

2. **Section 3.1.2: As you mentioned before, ATAL composition is dominant by sulfate, nitrate, ammonium and organics. Why does the organic decrease in Sep if ATAL materials still transport into ExLS region? Is it because the SOA from ASM is not high in k+?**

Thank you for raising this point. The decrease in organic matter in September does not contradict the transport of ASM-influenced material into the ExLS. Our interpretation is that the organic signal during the Early Phase was strongly affected by biomass burning, as indicated by enhanced organic mass concentrations together with a higher fraction of potassium-dominated particles. In contrast, the Late Phase shows a clearer enhancement of nitrate and ammonium, which we attribute to ASM-influenced outflow.

Therefore, the September increase of ASM-influenced material is mainly reflected in ammonium nitrate rather than in an increase of bulk organic matter. Organic aerosol associated with ASM outflow may still be present, but its absolute contribution appears to be smaller than the biomass burning related organic enhancement observed during the Early Phase. In addition, chemical aging, and possible loss of semi volatile organic material during transport and stratospheric residence may further reduce the organic mass contribution.

We would also like to point out that the reviewer is right in assuming that the SOA from the ASM is not highly enriched in K^+. This has also been confirmed by Appel et al. (2022) and Ebert et al. (2024). We also clarify that potassium-dominated particles are used here as an indicator of biomass burning influence, not as a marker for secondary organic aerosol from the ASM. Secondary organic aerosol formed by precursor gases from the ASM region is not expected to be necessarily enriched in K^+. According to Appel et al., 2022 and Ebert et al., 2024, organic matter in the ATAL is secondary organic aerosols without primary particle contribution from the near surface region. We also added this point to our manuscript.

L345: *"[...] This particle type should therefore not be interpreted as a marker for secondary organic aerosol associated with the ASM-influenced air mass. Previous observations within the ATAL indicate that organic matter in ASM-influenced air masses is mainly associated with secondary aerosol formation from precursor gases, whereas direct transport of primary particles from near surface sources is strongly limited by efficient wet scavenging during convective transport (e.g., Appel et al., 2022; Ebert et al., 2024). Consequently, ASM-related organic aerosols are not necessarily enriched in potassium components. Potassium-dominated particles are specifically used here as an indicator of BB influence. [...]"*

Specific points:

3. **Line 173: What causes the detection change between different flights?**

   We thank the reviewer for this important question. The exact cause of the varying ERICA-AMS detection limits between individual flights cannot be fully reconstructed retrospectively. However, several instrumental and operational factors likely contributed to the observed variability.

   First, before flight F08, the ERICA-AMS vacuum chamber was unintentionally re-pressurized with ambient air during ground-based operation due to a technical handling issue. Although the instrument was subsequently pumped down extensively and the vacuum conditions returned close to the initial operating conditions, we cannot fully exclude that this event slightly affected the instrumental background (in particular, the water vapor content in the chamber) and therefore the detection limits during the following flights.

   Second, during flight F09 the ERICA instrument encountered an intense biomass burning plume with exceptionally high aerosol concentrations. To protect the instrument, the inlet valves were temporarily closed during the measurement period. Although the system was continuously pumped afterwards, minor residual contamination effects inside the instrument cannot be entirely excluded and may have contributed to variations in the background signal and detection limits.

   Importantly, however, we would like to emphasize that the resulting detection limits remained within a reasonable range for UTLS aerosol measurements and did not affect the overall conclusions of this study. All averaged concentrations shown in this work remained significantly above the corresponding detection limits.

   L207-Supplement: *"A possible explanation for the variability in the ERICA-AMS detection limits (Tab. 2 in the main text) between individual flights is related to instrumental conditions during the campaign. Prior to flight F08, the ERICA-AMS chamber was unintentionally re-pressurized with ambient air during ground-based operation. In addition, during flight F09 the instrument sampled an intense BB plume with exceptionally high aerosol concentrations, which may have caused minor residual contamination effects despite subsequent pumping procedures. However, the instrument was extensively pumped after both events and returned close to normal operating conditions. The resulting detection limits remained within a reasonable range for UTLS aerosol measurements and did not affect the interpretation of the presented results."*

4. **Line 180: What's the NOAA standards?**

   We thank the reviewer for pointing this out. The term NOAA standards was not sufficiently precise in the original manuscript. In this context, it refers to calibration standards that are traceable to the NOAA calibration scales for trace gases, which provide internationally comparable reference scales for atmospheric measurements. The laboratory standards used for UMAQS were calibrated before and after the campaign against standards traceable to these NOAA calibration scales, including the WMO calibration scale for CO. We have revised the sentence to clarify this point and avoid ambiguity.

   Website: https://gml.noaa.gov/ccl/scales.html /last visit: 14-05-2026

L179: *"[...] Based on in-situ calibrations against secondary laboratory standards, which were calibrated before and after the campaign against standards traceable to the NOAA calibration scales (NOAA Global Monitoring Laboratory) for atmospheric trace gases to ensure global comparability, the total uncertainty (2 sigma, 1 Hz) is estimated to be 0.3 ppbv for N_2O and 1.4 ppbv for CO. [...]"*

5. **Figure 4: Figure right panel, add "Aug" and "Sep" after Early phase and Late phase. Caption second line "the Late and Early Phases (August and September)": Switch Sep and Aug to be consistent with Late and Early phases.**

   Done.

   Figure 4/5: *"[...] between the Late (late September) and Early (mid-August) Phase, [...]"*

6. **Line 382-383: This sentence is a little vague. I am not sure if you are talking about one mechanism or two here. The first sentence reads like a physical process that increase the H2SO4 percentage through coagulating NO+-rich particle with Sulfur-rich particle or condensing H2SO4 gas. Maybe adding "through chemical process" at the end of line 383 after "the depletion of nitrate".**

   Thank you for pointing this out. We agree that the original sentence was not sufficiently clear because it combined different processes in one formulation. Our intention was to describe two related but distinct effects during stratospheric aging of NO^+-rich particles: first, the relative enrichment of sulfate through the uptake of sulfuric acid by condensation and/or coagulation with sulfate-rich particles, and second, the depletion or replacement of particulate nitrate through chemical processing during longer residence time in the stratosphere. We have revised the sentence to make this distinction clearer.

   L382: *"[...] 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). [...]"*

7. **Line 448: I don't understand this "significant chemical processing"**

   Our intention was to refer to chemical aging during stratospheric residence time, reflected by changes in the internal mixing state of NO^+-rich particles. In particular, we refer to the relative enrichment of sulfate compared to nitrate, changes in the ammonium-to-nitrate signal ratio, and the shift from less oxidized to more oxidized organic ion signals.

   L447: *"[...] This further indicates that NO^+-rich particles can be observed in the stratosphere even after a few months. During the residence time in the stratosphere, their composition evolves toward more oxidized organic matter signatures and a relative replacement of nitrate by sulfate, indicating progressive particle aging. [...]"*

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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