

Replies to Reviewer # 2 Comments/Suggestions

General Comments:

The paper provides a comprehensive review of the key findings related to the Asian Tropopause Aerosol Layer (ATAL). It utilizes data from balloon measurements conducted at three different locations in India to construct mean profiles of aerosol backscatter, water vapor, ozone, and other parameters for the UTLS region. Additionally, the authors present and discuss the computed radiative forcings and heating rates based on three different aerosol scenarios for ATAL. Overall, the paper serves as a valuable review of the ATAL observed over the Indian region, although it is important to note that the full extent of ATAL is much broader.

Reply: *We sincerely appreciate the reviewer's thorough summary of our study. Based on the insightful comments and suggestions provided, we have made significant revisions to improve the manuscript.*

Specific Comments:

The major limitation of this study is the lack of detailed information on the size-resolved chemical composition of the ATAL. In the absence of this data, the authors propose three scenarios—sulfates, nitrates, and anthropogenic particles—to represent the composition of ATAL and calculate the corresponding radiative forcing and heating rates. Previous studies have investigated the radiative forcing of ATAL aerosols, focusing on components such as organic carbon, black carbon, and sulfates. However, in reality, ATAL is a mixture of all these aerosol types, and its composition varies from the outer to the central regions. Therefore, characterizing ATAL simply as consisting of nitrates, sulfates, or anthropogenic particles, as done in the present study, oversimplifies the actual complexity of its composition and the obtained radiative forcing and heating rates can be far from reality.

Reply: *We acknowledge the reviewer's concern regarding the simplified representation of ATAL composition in our initial analysis. In the revised manuscript, we now account for ATAL as a mixture of aerosol species—sulfates, nitrates, organic carbon, and ammonia—by incorporating composition estimates from recent modeling and observational studies. This approach better reflects the spatial variability and complexity of ATAL's composition, providing a more representative assessment of its radiative effects.*

Technical corrections:

L46: The geographic area of the ATAL is described as 10 to 40 deg N and 10 to 140 deg E in page 2 line 46 and in Fig 1 caption it is shown as 15 to 40 deg N and 15 to 105 deg E, whereas Gadanki is at 13.5 deg N which is however shown within the red box in the figure.

Reply: *We thank the reviewer for bringing this to our attention. We have corrected the figure in the revised manuscript.*

L131: it is mentioned that “These campaigns involved over a hundred balloon flights..” whereas Table 1 shows a total of 21 flights.

Reply: We appreciate the reviewer's observation. The campaign indeed included around a hundred balloon flights; however, for this study, we specifically selected only those that reached at least 20 km altitude to ensure continuous backscatter and atmospheric profiling. Many flights did not meet this criterion, as they were intended for other objectives, allowing only a few to float within the 16 km to 18 km range. Additionally, some flights lacked ozonesondes/COBALD sensors. Consequently, 21 flights were selected for our analysis. To avoid any ambiguity, we have revised this statement in the manuscript for better clarity.

Fig 4: ASY for nitrate shows a dip at 100% RH which is difficult to comprehend.

Reply: We thank the reviewer for bringing this to our attention. The dip in ASY at 100% RH was due to a technical plotting error, which has now been corrected. In the revised manuscript, we have refined the size distribution parameters for nitrate aerosols—mode radius ($0.15 \mu\text{m}$) and standard deviation ($2 \mu\text{m}$)—within the $0.5\text{--}2 \mu\text{m}$ size range, following Zhang et al. (2012) and Vernier et al. (2022). Additionally, we have revised the SSA and ASY calculations across different RH bins using the methodology from Zhang et al. (2012). Since the UTLS remains predominantly dry, we now present SSA and ASY values for nitrates under dry conditions (zero RH) across altitude bins in the updated manuscript.

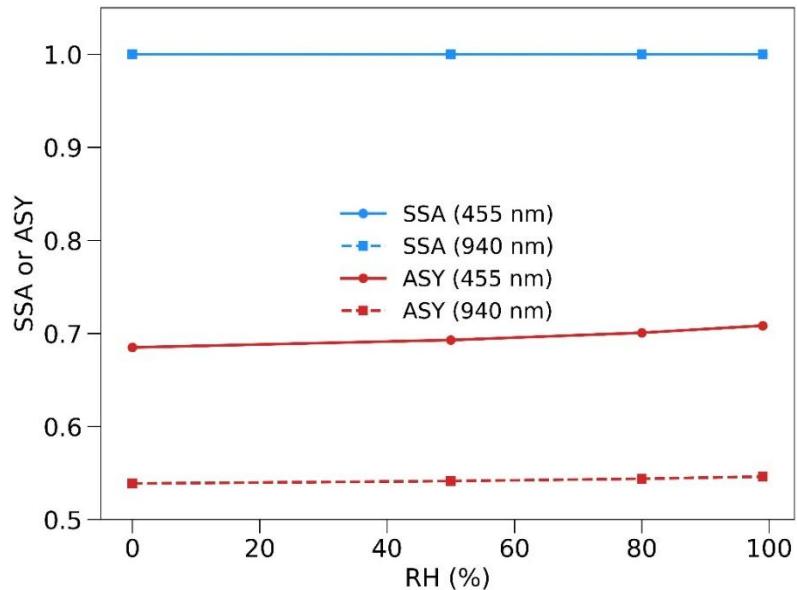


Figure R1: Single Scattering Albedo (SSA) and Asymmetry Parameter (ASY) for nitrate aerosols at selected Relative Humidity (RH) levels. In the revised analysis, SSA and ASY values for nitrates are considered under dry conditions (zero RH) due to the predominantly dry nature of the UTLS.

L181-184: It is mentioned that the measured water vapour values over Varanasi are higher than that over Hyderabad and Gadanki. However, it is difficult to appreciate this fact in Figure 2c, unless log scale is used for the concentration values.

Reply: We appreciate the reviewer's suggestion. In the revised manuscript, we have updated Figure R2 to use a logarithmic scale for water vapor concentration, making the differences

between locations more apparent. To maintain the manuscript's focus on the radiative impacts of UTLS aerosols, we have moved this figure to the supplementary section.

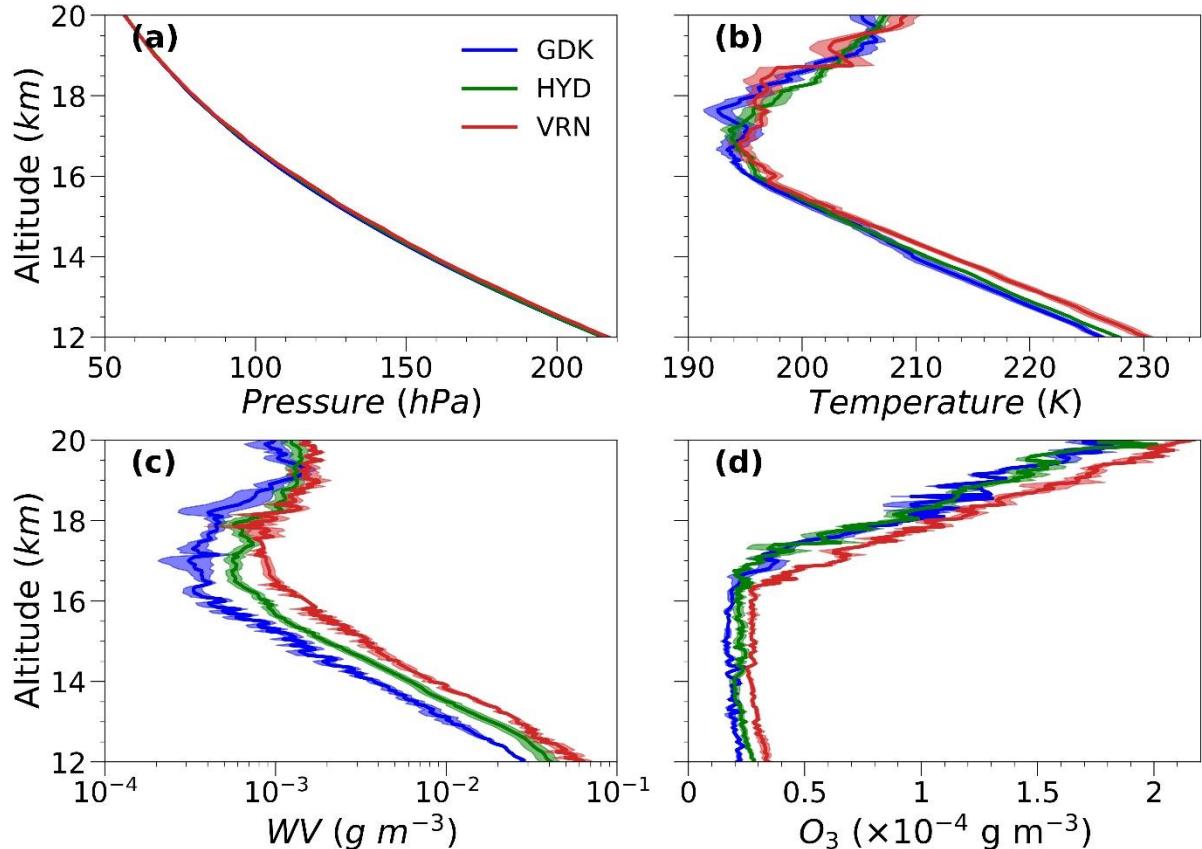


Figure R2: Vertical profiles of (a) Pressure, (b) Temperature, (c) Water Vapor Density (WV; log scale), and (d) Ozone Density (O_3) in the UTLS region for Gadanki (GDK), Hyderabad (HYD), and Varanasi (VRN). The log scale in panel (c) enhances the visibility of variations in water vapor concentration across locations.

Table 2: Aerosol types within boundary layer for Varanasi, Urban (60%) and Continental Average (20%) doesn't add to 100%.

Reply: We appreciate the reviewer for bringing this to our attention. This was a typographical error—‘Urban’ aerosols contribute 80%, not 60%. We have corrected this in the revised manuscript. Additionally, since the primary focus of the study is on UTLS aerosols, we have moved this table to the supplementary information.

L431: “13 km to 19 km” repeated twice.

Reply: We have corrected this in the revised manuscript.

Equ 16: Since DARFx is the difference between relatively two large numbers it can be within the uncertainty of the computed ARF.

Reply: *We agree with the reviewer's observation. To improve clarity, we have revised the notation from ΔARF_x to δARF_x , distinguishing it from the total forcing of a given layer. The computed δARF_x represents atmospheric forcing differences from the surface (0 km) to the top of the atmosphere (30 km). While δARF_x values reach up to 0.5 W m^{-2} , they remain smaller than the standard deviation of the total column ARF (up to 5 W m^{-2}), indicating that UTLS aerosol composition changes have a minor impact on total atmospheric forcing. However, within the UTLS region itself, δARF_x values range from 0.06 to 0.28 W m^{-2} , exceeding the standard deviations of UTLS forcing (0.01–0.08 W m^{-2}). This suggests that δARF_x in the UTLS is significant, highlighting localized but strong radiative effects.*

We sincerely thank the reviewer once again for valuable suggestions, which have significantly enhanced the content of the manuscript.
