Response to Reviewer -2 (egusphere-2026-3032)

General comment: This study utilizes a year-long surface O_3 measurement dataset from Leh in the Trans-Himalayan region, alongside satellite observations and model simulations, to analyze local O_3characteristics. The O_3observations reveal a unique double-peaked diurnal variation and strong daytime O_3enhancement compared to other high-altitude sites. WRF-Chem simulations suggest that this region primarily serves as an O_3sink, with O_3levels strongly influenced by complex local circulation and transport processes. In addition, analysis of large-scale model outputs indicates that O_3over Ladakh is mainly contributed by background CH_4 oxidation, stratospheric intrusions, and emissions from South Asia and other surrounding regions. Overall, this study provides a comprehensive analysis of O_3dynamics and source attribution in the Ladakh region, which fits well within the scope of Atmospheric Chemistry and Physics. However, there are several weaknesses and areas requiring further clarification and justification. Therefore, I recommend a major revision for this manuscript.

Response: We thank the reviewer for providing detailed comments and suggestions, which have helped greatly in improving the manuscript. We have addressed all the comments positively and incorporated all the suggestions. Point-by-point responses are given below. Line number mentioned here are corresponding to the manuscript file in track-change mode. Note that a minor correction factor, which was missed previously, is applied to the in situ data which has increased the ozone values slightly (~1-1.5 ppbv) and figures/text are revised accordingly. This does not change any conclusions.

Major Comments

Comment#1: Section 3.1: The distinct double-peak O_3diurnal pattern shown in Figure 2 may be closely linked to complex local mountain meteorology (e.g., upslope/downslope winds, valley breezes, or localized precipitation). Are there additional meteorological observations or nearby weather stations in the Leh region that could be analyzed to elucidate the interaction between O_3variations and local wind dynamics? Furthermore, is the WRF-Chem model capable of capturing these fine-scale circulation patterns at the current grid resolution? The authors should provide further discussion and justification on these aspects.

Response#1: Yes, mountain-valley wind systems and vertical mixing etc. could have role in governing the observed diurnal patterns. As raised by the reviewer and discussed in introduction of the paper, the in-situ observations are not available from this region, motivating us to set up a representative station. Nevertheless, we have explored the association between ΔO_3 and wind direction (new figure 4) using 1-week long observations. The analysis indicated that the lower O_3 values in the morning are associated with wind direction 0–60° N, whereas the higher values during noon to late night hours are associated with wind from 90–315° N (Northwest, southwest and southeast sectors). Additional analysis (new figure 5) showed higher concentration of precursors (NO_2 and CO) in 90–315° N sector and hence explaining noontime build-up to an extent. (Line# 362-385)

We have applied WRF-Chem model at higher resolution (12 km × 12 km), as compared to several previous modeling studies over South Asia. We do agree with the reviewer that even then the dynamics may not be resolved adequately as the topography is very complex. Unfortunately, the in-situ atmospheric measurements remain quite scarce in the region, which has been to a great extent the main motivation to initiate this study. Lack of systematic measurements from different stations limits a deeper evaluation of model meteorology/dynamics as of now. Nevertheless, a quick look at the vertical wind (figure R2_1) indicates prevalence of mountain-valley wind system in the model,

somewhat weaker than seen in other part of Himalaya using same model (Sarangi et al., 2014). The likely role of biases in dynamics at fine-scales is acknowledged more clearly in the revised version of the paper (Line # 341-342).

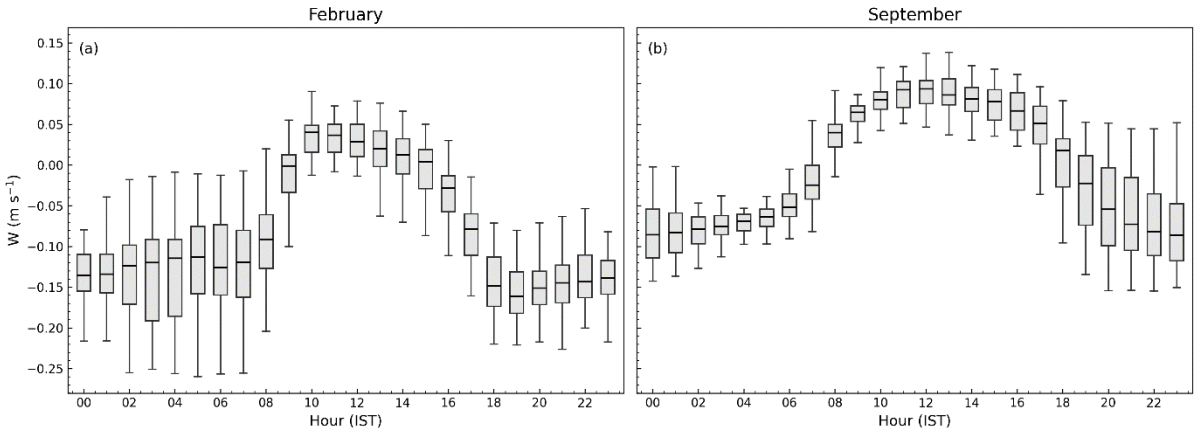


Figure R2_1: Diurnal variation in the vertical wind over Leh during February and September 2025 derived from the WRF-Chem simulation.

Comment#2: Section 3.4: Using the satellite-derived HCHO/NO_2 ratio (FNR) as an indicator for the surface O_3formation regime over Ladakh comes with significant uncertainties. Both TROPOMI NO_2 and HCHO retrieval products contain intrinsic systematic biases, and TROPOMI HCHO is subject to substantial retrieval noise (low signal-to-noise ratio), particularly over pristine, high-altitude environments. Taking the ratio of these two products can severely compound these errors. While retaining this analysis is acceptable, the authors should explicitly discuss the limitations of using satellite FNR in this remote region. Additionally, I suggest providing spatial distribution maps of the multi-year or seasonal mean TROPOMI NO_2 and HCHO columns over Ladakh so readers can assess data quality and signal levels.

Response#2: We have mentioned the limitation of using satellite-derived HCHO/NO_2 ratio (FNR) for inferring the chemical regime of surface O_3 based on literature. As the reviewer has also pointed out, there are several factors contributing to the uncertainties. In particular, (a) greater noise under low concentrations over mountains, (b) inherent chemistry assumption, (c) column-to-boundary layer translation, and (d) limited spatial representativeness due to coarser resolution of satellite. Previous studies have reported positive biases in TROPOMI HCHO and NO_2 for cleaner regions, in contrast to negative biases over polluted/urban regions. These points are discussed along with references for uncertainties in the revised version (Line# 477-487).

As suggested, we have also analysed multi-year average climatologies of NO_2, HCHO and CO using TROPOMI observations during October 2018–September 2026 (new figure 5; Line# 370-378). This is in line with our results that precursors (NO_2 and CO) are higher over valley regions. The emissions from nearby valley, in the proximity of the study site, could influence the diurnal or day-to-day changes in the observed O_3.

Minor Comments

Comment#3: Lines 201–204: Given the complex topography over Ladakh, applying meteorological grid nudging (especially for horizontal winds) within the planetary boundary layer (PBL) can distort

or artificially suppress fine-scale local circulations simulated by WRF dynamics. It is generally recommended to nudge only above the PBL height. Could the authors also clarify specify which reanalysis dataset (e.g., ERA5, NCEP FNL) was used for nudging?

Response#3: We agree with the reviewer that nudging within PBL may have some suppressing effects on the local circulations computed by WRF. Nevertheless, we had kept nudging active to ensure that on larger-scales our dynamics should be consistent with the ERA5 reanalysis. Few comprehensive evaluation studies of WRF-Chem over South Asia had applied stronger nudging (coefficient ~ 6×10^{-4}) at all vertical levels (Kumar et al., 2012a, b) showing robust agreements with observations. Whereas, we have used rather moderate nudging (3×10^{-4}) for the reasons stated by the reviewer. We have included information on the data used (i.e., ERA5) and the rationale for nudging more clearly in revised version (Line# 225-227).

Comment#4: Lines 205–206: The anthropogenic emission inventory used here (base year 2015) lags the target simulation period (2024–2025) by nearly a decade. Since the authors state that the Ladakh region has experienced rapid urbanization and increasing tourism, have they evaluated how these emission changes might impact model performance and source attribution?

Response#4: We agree with the reviewer's observation regarding the rising anthropogenic emissions over study region. Unfortunately, up-to-date and accurate inventories, accounting for increase in various local/regional activities in and around Trans-Himalaya remain lacking. Analysis of the recent EDGAR inventory (Figure R2_2) suggests large increase (50–60%) in 2015 across South Asia, as compared to 2005. However, the enhanced emissions tend to sustain or increase only slightly (10–20%) in 2022. Somehow, over the Trans-Himalayan region, inventory indicates decline towards 2022. We have used 2015 inventory which considers overall large increase whereas the decline in emissions over study region towards recent years could be very uncertain. This uncertainty could be mainly due to lack of new local activity data from this region.

Here, we use WRF-Chem simulation only to infer key processes influencing the diurnal and day-to-day ozone variability. The main results from WRF-Chem are shown in terms of ΔO_3 rather than in absolute values. We do realize that emissions remain uncertain and therefore relied fully on in-situ measurements while inferring ozone impact on human health and vegetation. The development and evaluation of inventories is critical for quantifying the role of changing local emissions, which will be attempted in future once measurements of O_3 precursors become available. The uncertainties/limitations of this study, along with future perspectives are stated more clearly in the revised manuscript (Line # 248-252).

Figure 12, showing the trend in O_3 and its sources, includes impacts of changes in emission, however, only up to 2018. For the later time period, one would speculate similar behaviours as shown in figure 12, however further simulations with updated inventories would be required for better insight on source attributions for the recent time.

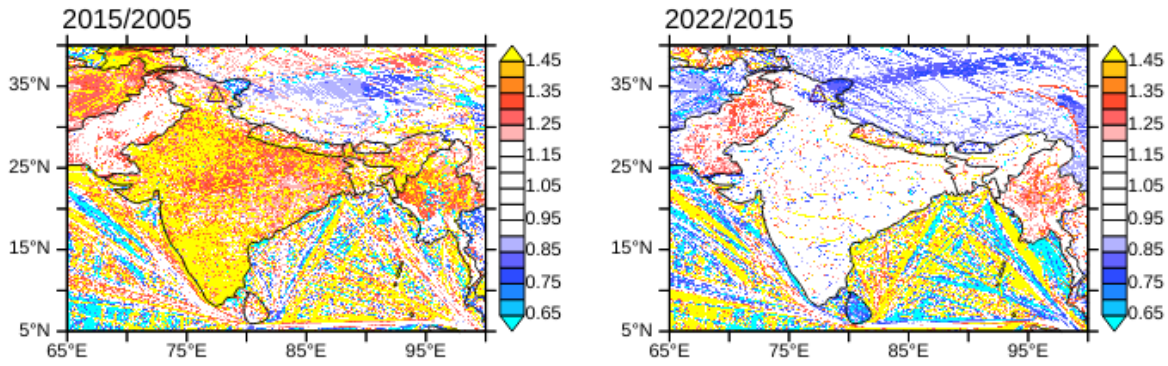


Figure R2_2*: Ratio of anthropogenic NO_x emissions estimated from the EDGAR inventory. Left: 2015 to 2005, and Right: 2022 to 2015.*

Comment#5: Section 3.1: The observed double-peak diurnal pattern in O_3 is absent in certain months (e.g., afternoon dips do not occur in December, January, and May). Could the authors provide more explanation for these seasonal differences?

Response#5: Mountain-valley circulation (and entrainment) could play important role as indicative from the association between O_3 and wind direction (new figure 4 and related discussions). Unfortunately, we do not have co-located measurements of winds and boundary layer height for deeper analysis. WRF-Chem simulations at spatial resolution of 12 km × 12 km may not fully resolve the dynamics due to complex topography (Line# 341-342), which is also visible in Fig 3b as model does not capture the observed double-peak. Therefore, at this stage, we are only reporting this new feature, which has not been seen in e.g. central Himalaya. The explanations would be attempted in future studies through co-located measurements related to atmospheric dynamics and improvements in the model.

Comment#6: Line 318: The phrase "photochemical loss" reads awkwardly in this context; consider using the standard term "net chemical loss" or simply "chemical loss."

Response#6: We have used “chemical loss” now (Line# 345).

Comment#7: Line 340: The chemical tendencies reflect the daily variation of the chemical term rather than the total net budget of O_3. In addition, the magnitude of the chemical tendencies shown in Figure 3d appears unusual—one would expect it to oscillate around a balanced baseline. Please double-check these values and clarify the budget definition.

Response#7: Yes, chemical tendency represents change in O_3 only due to chemical process, not the total net change in O_3. The unit of tendencies shown in figure 3d was mistakenly written as “pptv h-1”, which is corrected as “ppbv h-1” and hence the magnitudes of tendencies are reasonable with corrected unit. The tendencies in the unit of ppbv h^{-1} (difference between consecutive values of cumulative tendency) at hourly interval oscillate around a baseline reflecting embedded diurnal changes and sum of all tendencies (conv_ten, chem_ten, vmix_ten, and adv_ten) represent hourly net change in O_3. Since our aim is to analyse changes on day-to-day scale, tendencies shown in figure 6 are at daily scale (ppbv d^{-1}) and hence diurnal oscillations are not seen. Daily tendencies (i.e., cumulative or total change in O_3 over a day due to a given process) are obtained by subtracting values of cumulative tendencies at 13 h on $(n-1)^{th}$ day from value at the same time on n^{th} day. We have double-checked the values and verified that these are correct. (See Line# 391-399)

Comment#8: Section 3.4: Please clarify how the TROPOMI NO_2 and HCHO retrieval data were processed (e.g., spatial/temporal averaging windows). Were the satellite observations sampled strictly over the co-located ground site pixel or averaged over a wider regional domain?

Response#8: For the estimation of chemical regime of O_3 (figure 8), single pixel strictly co-located with ground-site is considered. Temporal window is ±15 minutes. However, for the estimation of trend, shown in figure 13, a bigger region including nearby town in proximity of the study site (77.41°–77.70° E; 34.08°–34.23° N) is considered to include larger number of observations for better monthly representativeness. This is mentioned in the revised manuscript (Line # 462-463 and 661).

Comment#9: Lines 424–429: Could the authors comment on why the model indicates the strongest stratospheric O_3 intrusion in June, even though the vertical velocity is relatively low during this period?

Response#9: Analysis of spatial distribution indicates that instead of direct downward stratospheric intrusions, ozone is horizontally transported to the study region in June (Fig R2_3). Liao et al., 2025 have also reported intrusions over Tibetan Plateau during June which could be transported horizontally, influencing other high-elevation regions. Nevertheless, further studies would be needed to better understand the role of stratospheric intrusions. This is mentioned in the revised manuscript (Lines# 504-507)

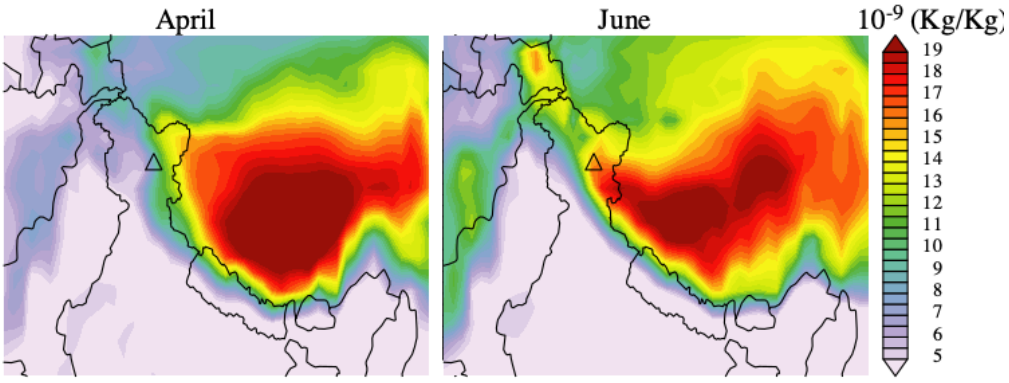


Fig R2_3: Mean distribution of O3s, derived from CAMS reanalysis for April and June 2025.

Comment#10: Figure 7c: Please clarify the mathematical definition and calculation methodology used to determine "O3 build-up."

Response#10: O_3 build-up in figure 9 is same as in the figure 2. It is defined as the noontime maximum minus morning minimum, which is mentioned in figure caption (Line# 514)

Comment#11: Lines 458–459: Please explicitly list the station names, coordinates, or mark their locations on a regional map.

Response#11: The stations are marked in figure 1a (Line# 534-535).

Comment#12: Figure 8a: Please clarify how the dashed line was derived.
Response#12: Dotted line is derived by simply connecting pre-monsoon to post-monsoon months. Pre-monsoon month is chosen just before sharper monsoonal decline (April, May or June) and post-monsoon month (September, October or November) is chosen as corresponding to the peak after the monsoonal trough (Line# 548-551).

Comment#13: Line 472: Should "2024-2015" be "2024–2025"?
Response#13: Yes, it is 2024–2025. Corrected now (Line# 555).

Comment#14: Section 3.6: Please provide a brief description of how O3 source attribution is estimated in the CAM-Chem simulations.
Response#14: We have added a brief description in section 2.4 (Line # 190-212).

Comment#15: Figure 9a: Please clarify how the CAM-Chem model output was combined with stratospheric O3 estimates from CAMS reanalysis, as this methodology is currently omitted from the text.
Response#15: Both, CAM-Chem and CAMS, provide surface O_3 and stratospheric O_3tracer (O_3s). To incorporate CAMS-O_3s in CAM-Chem results, we have subtracted CAM-Chem estimated O_3s and added CAMS derived O_3s (Line# 187-189).

Comment#16: Section 3.7: Given that emissions from other regions contribute substantially to O3 levels over Ladakh, have the authors evaluated which specific upstream source regions show the largest transport impact?
Response#16: We have analysed contribution from different regions and revised the figure. In NO_x perspective, contributions from outside South Asia are as follows. East Asia: 4–12% (2–7 ppbv), Middle East: 3–10% (2–6 ppbv), Ocean: 2–8% (1–4 ppbv), North America: 3–5% (2–3 ppbv), Europe: 1–4% (1–2 ppbv), and Rest of the world: 8–11 % (5–6 ppbv). (Line# 600-610)

In VOC/CO/CH_4 perspective, 6–8 ppbv is contributed by the regions outside South Asia. Contributions from East Asia (4–5%; 2–3 ppbv), Middle East (0–2%; 0–1 ppbv), and North America (1–2%; ~1 ppb) remains smaller. (Line# 617-621)

This suggest that the contributions of NO_x emissions from East Asia and Middle East are higher over the Trans-Himalaya. Interestingly, the contribution of NO_x emission (17–21 ppbv) is much more than VOC/CO contributions (6–8 ppbv) from the regions outside South Asia (Line# 724-726).

Comment#17: Lines 570–580: The observed multi-year increases in TROPOMI NO2 and HCHO columns could also be driven by upwind emission trends across South Asia (via regional transport) or climate variability (e.g., warmer temperatures enhancing biogenic VOC and soil NOx emissions). Do the authors have access to local emission datasets, traffic statistics, or visitor numbers to substantiate the claim that these satellite trends are primarily driven by local anthropogenic activities?

Response#17: Though we do not have consistent data on emission, traffics or number of tourists, followings based on the literature are the supporting evidences for the increase in local/regional anthropogenic activities. (a) Number of tourists in Ladakh have increased over the recent years: 0.28, 0.33, 0.28, 0.52, 0.38, and 0.33 millon in 2017, 2018, 2019, 2023, 2024, and 2025, respectively (https://www.ijmra.us/project%20doc/2020/IJRSS_DECEMBER2020/IJRSS12Dec20-21086.pdf, https://www.ibef.org/states/ladakh-presentation, https://lokniwas.ladakh.gov.in/press-details/369). (b) Satellite (Resourcesat-2 LISS-IV) based estimation of in build-up area over Leh town (77.55–77.63° E; 34.08–34.20° N) shows increase from 1249.9 h (12.5 km^2) in 2016 to 1755.3 h (17.5 km^2) in 2025 (https://vedas.sac.gov.in/vapps/lama/main.html?app=urban; https://vedas.sac.gov.in/vapps/lama/assets/LAMA-interim-report.pdf#page=16). This shows that anthropogenic activities are rising over the Trans-Himalaya and the increasing trend seen in the satellite observations has contribution of local emissions, besides the regional emissions. (Line# 680-690)

References:

Sarangi, T., Naja, M., Ojha, N., Kumar, R., Lal, S., Venkataramani, S., Kumar, A., Sagar, R., Chandola, H.C., 2014. First simultaneous measurements of ozone, CO, and NOy at a high-altitude regional representative site in the central Himalayas. J. Geophys. Res. Atmos. 119 (3), 1592–1611. https://doi.org/10.1002/2013jd020631.

Kumar, R., Naja, M., Pfister, G. G., Barth, M. C., and Brasseur,G. P.: Simulations over South Asia using the Weather Researchand Forecasting model with Chemistry (WRF-Chem): set-upand meteorological evaluation, Geosci. Model Dev., 5, 321–343, https://doi.org/10.5194/gmd-5-321-2012, 2012a.

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Liao, Z., Zhang, J., Gao, M., and Ma, Z.: Widespread stratospheric intrusion influence on summer ozone pollution over China revealed by multi-site ozonesonde and validated EAC4 reanalysis, Atmospheric Chem. Phys., 25, 14865–14877, https://doi.org/10.5194/acp-25-14865-2025, 2025.