Response to Referee #2 (Dr. Juseon Bak)

We thank the reviewer for their time and for these helpful comments. We carefully considered all your comments and made every effort to address them in this revised manuscript. In this response letter, we have repeated each reviewer's comments and provided our responses individually. Our responses are marked in blue. We also used "R2" to indicate Reviewer #2's comments and "A2" for our responses.

Reviewer 2

R2-1: The authors present an interesting and comprehensive analysis of sources and trends over western North America using a combination of source-receptor relationships, emissions information, and chemical model simulations. The manuscript addresses an important scientific question and provides valuable insights into the processes influencing ozone over the region. Overall, the study is well organized, clearly written, and fits well within the scope of ACP.

The comments below are intended primarily to improve the clarity of the interpretation and presentation.

A2-1: Thank you for your positive and thoughtful review. We have revised the manuscript based on your suggestions, and our detailed responses are shown below.

R2-2: Figure 2: For clarity, I suggest showing the same regional boxes in all panels. In addition, the figure could be improved to better highlight the spatial characteristics of the SRR fields. The map panels are highly compressed and important features are difficult to discern. I suggest increasing the map size and adding latitude/longitude grid lines or labeled latitude ticks. In addition, the PDFs occupy a substantial fraction of the figure and may be more effectively presented in a separate figure. This would allow the spatial patterns and source-region contrasts to be displayed more clearly.

A2-2: Thank you for the suggestion. As recommended, we have included the regional boundary boxes across all panels, increased the overall size of the maps, and added latitude and longitude grid lines. Additionally, to improve readability and ensure a clearer interpretation of the spatial data, we have removed the PDF plots from the previous version of manuscript.

Below is the revised plot shown in Fig. R1.

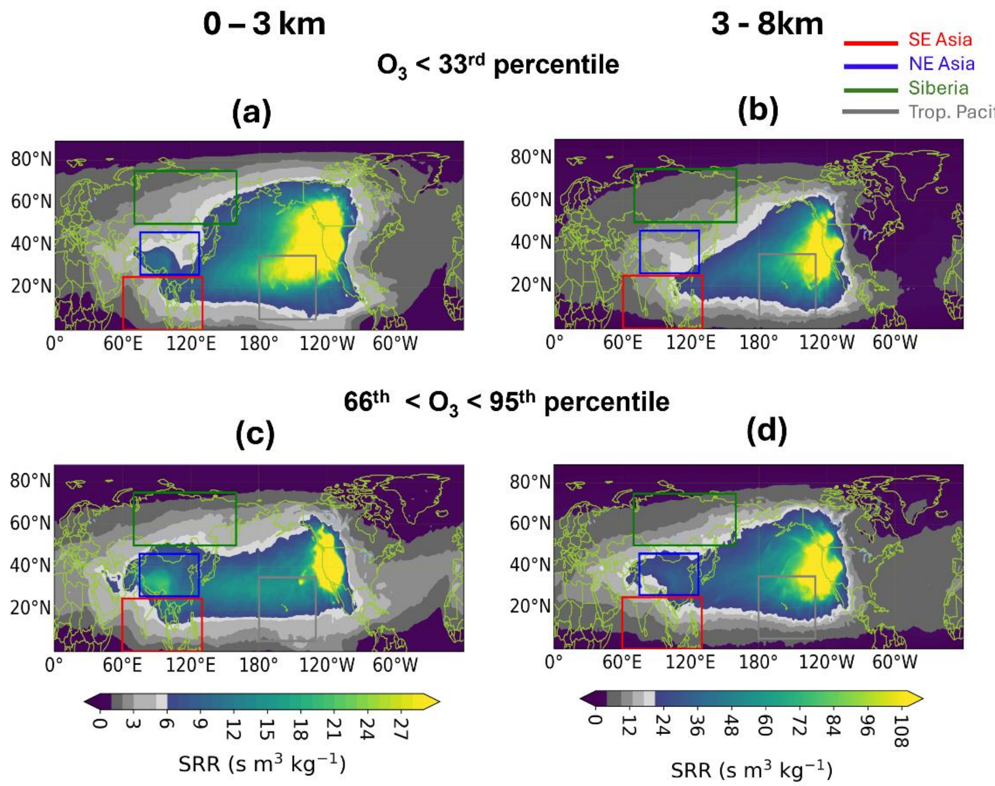

Figure 2 (in the revised manuscript). Maps of the source-receptor relationship (SRR): (top) lowest O_3 (< 5th percentile), (middle) low O_3 (< 33rd percentile), and (bottom) high O_3 (66th – 95th percentile) over the WNA for (a, c, e) 0–3 km level and (b, d, f) 3–8 km. The red, blue, green, and gray boxes in (a) represent SE Asia (0–25°N, 60–130°E), NE Asia (26–46°N, 75–127°E), Siberia (50–75°N, 70–160°E), and Tropical Pacific (5–35°N, 180°E–130°W), respectively.

R2-3: The term "latitudinal PDF" need to be revised. The presented PDFs appear to represent the distribution of SRR values for different source regions, rather than those as a function of latitude. Although the selected regions occupy different latitude bands, the PDFs are calculated over predefined geographic boxes with noticeably different longitudinal extents.

A2-3: Thank you for pointing this out. We agree that the original phrasing could be confusing. Because we removed the PDFs in response to the previous comment, this wording has now been removed as well.

R2-4: The statement "aligned specifically with the southern regions of East Asia and the tropical Pacific f 2a; SRR is higher in the red and gray boxed regions " does not appear to be fully supported by Fig. 2a. In particular, the PDF peaks for SE Asia are not clearly distinguishable from those of NE Asia or Siberia. While the Tropical Pacific exhibits a broader SRR distribution peaking at the highest SRR value, the SE Asia PDF does not obviously show either a uniquely high peak or a broader spread compared to the other source regions.

A2-4: We fully understand your concern regarding the differentiation between the source regions.

Regarding the modeling setup, LPDMs can occasionally assign unrealistically high SRR values to grid cells in the immediate vicinity of the receptor due to uncertainties in turbulent mixing and dilution; we therefore interpret these local maxima with caution. Beyond the grid cells adjacent to the receptor, we observe consistently elevated SRR over the West Coast and the tropical Pacific. This spatial pattern reflects actual atmospheric transport pathways during the 15-day backward simulation rather than a modeling artifact. Specifically, large-scale atmospheric circulation (e.g., jet exit regions) and topographic influences cause air masses to linger in these regions, resulting in longer residence times and higher SRR.

However, a high SRR in a given region does not automatically translate to a large contribution to downwind ozone. Ozone enhancement also requires the presence of precursor emissions and favorable atmospheric chemistry (net production or loss) along the transport trajectory. As shown in Figures 7 and S6 of the revised manuscript, both GMI O_3 and net O_3 production increase rapidly in the troposphere over Southeast (SE) Asia, Northeast (NE) Asia, and the tropical ocean.

Additionally, the PDF peaks for SE Asia and NE Asia are not completely distinct in the annual average, partially because of strong seasonal variations in transport driven by the shifting location of the jet stream (Fig. S8). While Fig. 2 illustrates the annual mean SRR, thereby blending this seasonal variability, the spatial distributions in Fig. 2 still reveal a clear contrast in the high-value SRR footprints over SE Asia and NE Asia when comparing low-ozone (< 33^{rd} percentile) versus high-ozone (66^{th} < O_3 < 95^{th} percentile) conditions. This spatial distinction is the primary takeaway we intend to highlight.

As you correctly noted, the SE Asia PDF does not exhibit an overly unique peak or a vastly broader spread relative to all other domains. Based on these features, we removed the PDF plot in the revised manuscript.

R2-5: The statement "Conversely, elevated O_3 concentrations are primarily associated with air masses transiting through NE Asia" appears stronger than what is supported by Fig. 2c. Although the NE Asia PDF exhibits a relatively higher peak, the difference does not appear sufficiently large. In addition, the Tropical Pacific shows a broad SRR distribution with a pronounced long tail, but SRR maps also do not clearly indicate a dominant contribution from any single source region. More generally, the contrast between Figs. 2a and 2c seems to reflect differences between maritime transport pathways and continental source influences. The relationship between lower-tropospheric O_3 over WNA and Asian continental source regions is certainly important. However, it may be difficult to directly infer this linkage from the backward trajectory results shown in Fig. 2 in the revised manuscript.

The Tropical/North Pacific region appears to be more directly connected to WNA than the Asian continent itself. Therefore, it may be more appropriate to first identify the Pacific regions that exert the strongest influence on WNA O_3 and then investigate the upstream source regions contributing to those Pacific air masses. Such an approach may provide a more robust framework for assessing the influence of Asian outflow on WNA ozone (0-3 km).

A2-5: Thank you very much for this thoughtful review. We agree with your observation that while the NE Asia PDF in the original manuscript exhibits a relatively higher peak, the difference is not particularly pronounced. We also agree that the SRR maps do not point to a single dominant contribution from any source region identified independently in this study. As mentioned in A2-4, the SRR represents a stochastic transport analysis. A larger SRR over the tropical/North Pacific does not necessarily imply a larger contribution to ozone unless there are corresponding emissions or chemical production associated with those regions.

As you noted, pollution transport is strongly modulated by large-scale meteorology, such as high- and low-pressure systems associated with atmospheric circulation and the jet stream, and its impact on WNA is intrinsically linked to trans-Pacific maritime transport. Hence, both high- and low-ozone transport events are heavily influenced by a mixture of maritime pathways and continental source signatures.

We carefully considered your suggestion to conduct an additional experiment. However, we anticipate that it would yield similar qualitative results, though specific features might vary depending on ozone levels or the season.

A comparable experiment was previously conducted by Ryoo et al. (2017) using 10-day kinematic back-trajectories, motivated by aircraft observations of elevated O_3 levels over the Pacific Ocean near the U.S. West Coast (see Fig. R2), where no clear local source was identified. Their study demonstrated that elevated O_3 events during specific episodes in May and June are highly associated with complex trans-Pacific air masses originating from the upper troposphere lower stratosphere (UTLS), the mid-troposphere, and the Asian lower troposphere. Specifically, their findings for these boreal spring episodic events showed that:

1. Elevated O_3 in the lower troposphere (1–4 km) originates from highly complex sources, with only minor influences from the North American boundary layer and the stratosphere.
2. The **primary O_3 transport** pathway involves **air masses originating from the Asian boundary layer that subsequently mix with mid-tropospheric air during trans-Pacific transit.**

This analysis demonstrates that no single source region is entirely dominant; rather, it is the mixing of distinct air masses during long-range transport that drives the overall ozone distribution observed over the Pacific Ocean. Given these transport patterns and dynamics in the region, we believe the current scope of our analysis sufficiently captures the complex, mixed nature of these transport pathways across varying ozone levels. We have revised the text in the manuscript to better articulate these points.

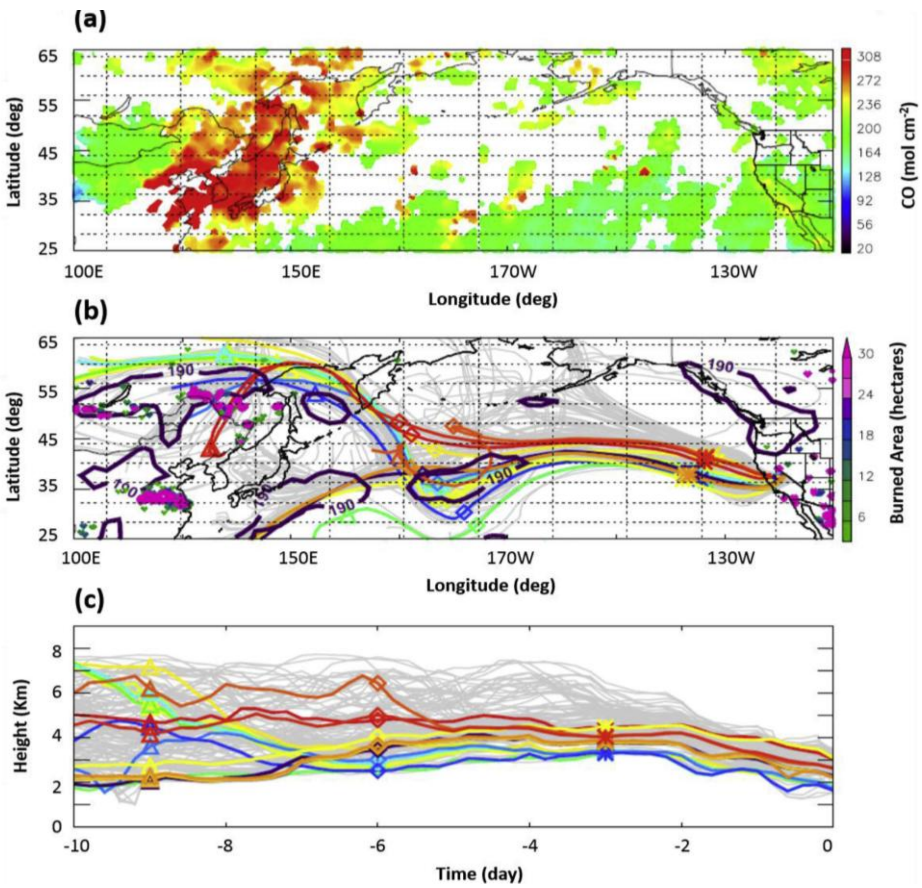

Figure R2. (a) Daytime column CO averaged over 21–30 May 2012 obtained from MOPITT data, (b) horizontal pathways, and (c) vertical evolution of 10 day back-trajectories releasing at 305, 308, 310, and 312 K (1.5–4 km) using MERRA-2 reanalysis data coming to California (36–42 °N, 125–118 °W on day 0 (30 May 2012 at 18 UTC)). Different colors of the trajectories represent the individual trajectories starting at locations closest to the AJAX flight path (37–37.5 °N, 124–120 °W) ending in different regions. The line colors closer to red (blue) signify the trajectories originating further north and higher altitude (south and lower altitude). The symbols represent the parcel locations at −3, −6, and −9 days. The colored regions in (b) represent burned area obtained from GFED. The purple contours are low OLR (190 W m^{-2}) at day −9, which is used as a proxy of deep convection. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Similar results are shown in Cooper et al. (2010), especially in springtime, as shown in Fig. R3.

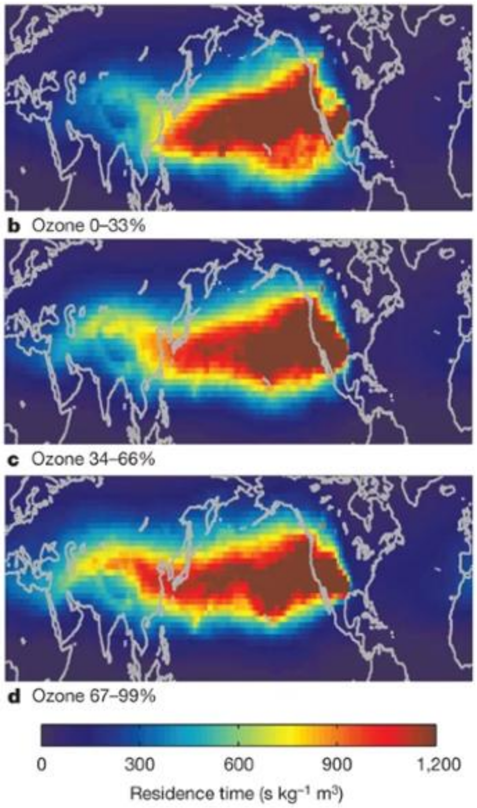

Figure R3. The average 1984–2008 retroplume for three ranges of ozone measurements, expressed as column residence times. b, 0–33rd, c, 34th–66th; d, 67th–99th. The figure is adapted from Cooper et al.(2010).

To address your concern, we have incorporated this additional explanation and context into the revised manuscript.

"Figure 2 illustrates the SRR maps for low (<33rd percentile) and high (66th-95th percentile) O_3 concentrations across the lower (0–3 km) and middle (3–8 km) troposphere. SRR maps are intrinsically linked to trans-Pacific maritime transport and are heavily influenced by a mixture of maritime pathways, continental air masses, and source signatures (Ryoo et al., 2017). From the map, we identify distinct transport corridors associated with varying O_3 levels. In the lower troposphere (0–3 km), low O_3 air masses exhibit high SRR values, with their peaks and dispersions aligned specifically with the southern regions of East Asia and the tropical Pacific (Fig. 2(a, c); SRR is higher in the red and gray boxed regions on the map). Conversely, elevated O_3 concentrations are primarily associated with air masses transiting through NE Asia (Fig. 2c).

This latitudinal contrast is further amplified in the free troposphere (3–8 km). While SRR values are generally larger in this layer compared to the lower troposphere, the spatial patterns remain consistent (Figs. 2b, d). For lower O_3 percentiles, air parcels predominantly traverse the tropical eastern Pacific and SE Asia. In contrast, higher O_3 percentiles are linked to air masses with extended SRR in the midlatitudes (15–45^{o}N), exhibiting transport pathways that originate or pass through NE Asia. Similar patterns were reported by Cooper et al. (2010), who showed distinct transport features corresponding to different O_3 levels, particularly in springtime. These characteristic transport dynamics are also evident on a seasonal basis (Fig. S4), confirming that the geographic origin of the air mass is an important factor of O3 variability over WNA."

R2-6: The seasonal results presented in Fig. S4 appear to contain additional information. It would be useful to briefly summarize the major seasonal differences in the main manuscript,

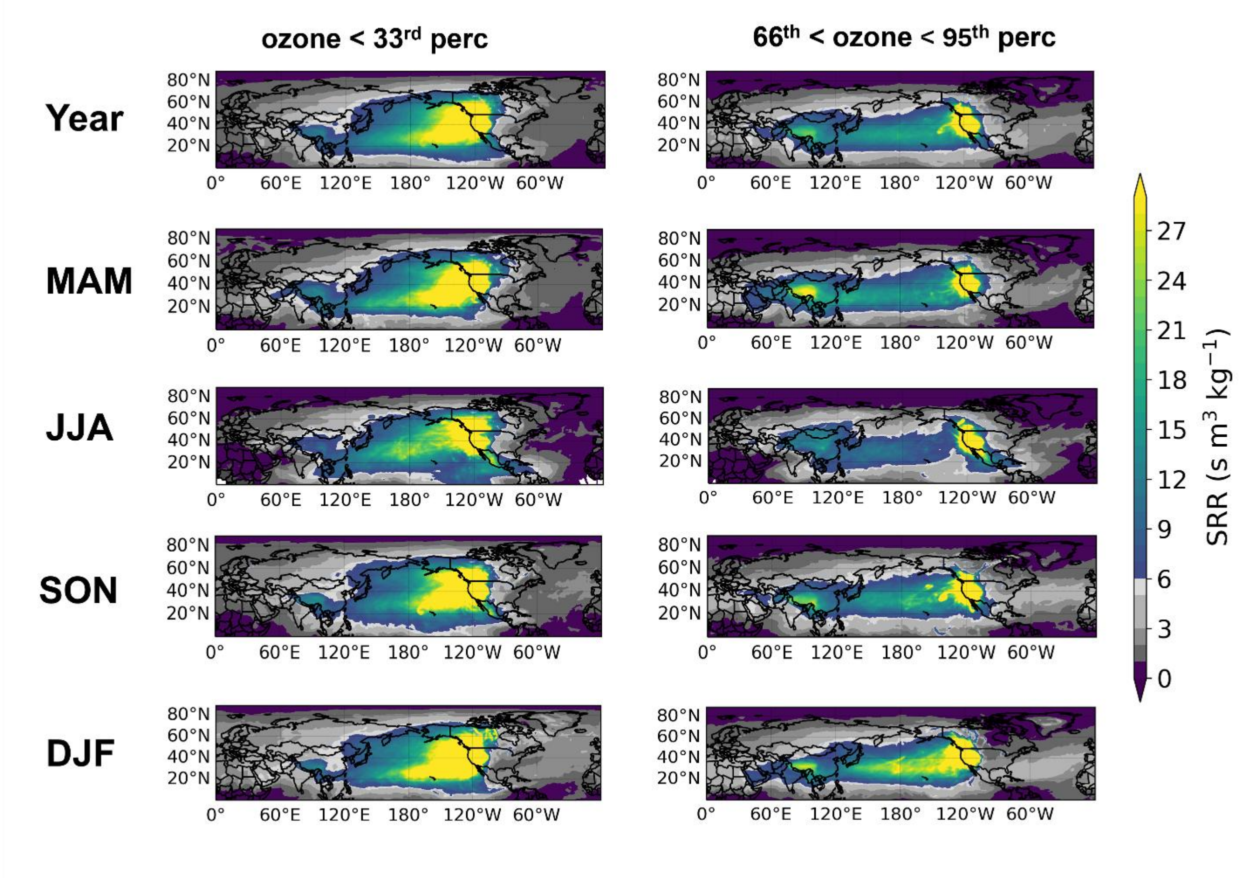

Figure S4. FLEXPART-ERA5 Source-Receptor Relationships (SRR, s m^3 kg^{-1}) for (left) low O_3 (< 33rd percentile) and (right) high O_3 (66th – 95th percentile) over WNA at the 300 m–3 km level, shown for (from top to bottom) the entire year, MAM (March, April, and May), JJA (June, July, and August), SON (September, October, and November), and DJF (December, January, and February), respectively.

A2-6: Thank you for this suggestion. We agree that expanding the description of the primary seasonal differences in the main manuscript adds valuable clarity. To address your concern, we have incorporated a detailed explanation of these seasonal variations into the results (3.1) section of the revised manuscript.

"*These characteristic transport dynamics are also evident on a seasonal basis: transport patterns weaken slightly and become more localized over WNA during boreal summer (June–August; JJA), whereas they strengthen during boreal spring (March–May; MAM) and boreal winter (December–February; DJF) (Fig. S4).*

This seasonal behavior aligns with findings by Ansari et al. (2025), who utilized explicit NO_X and VOC emission tagging to demonstrate an increasing influence of foreign anthropogenic NO_X and, to a lesser extent, global shipping NO_X, over the northwestern United States between 2000 and 2018. Similarly, Li et al. (2023) used the same explicit *dual-tagging technique and found increasing contributions of Asian NO_X emissions to Western U.S. O_3, particularly during winter from 1995 to 2019, further supporting our analysis. Taken together, these findings confirm that the geographic origin of an air mass is a key driver of O_3 variability over WNA."*

R2-7: Figure 3: The discussion of the increasing trend in the 8–13 km layer is interesting and well-motivated. Given that this layer overlaps with the UTLS region, it may also be worthwhile to briefly mention the potential role of stratosphere-to-troposphere exchange (STE), even if a detailed assessment is beyond the scope of the present study.

A2-7: As noted, an increasing trend in the UTLS source-receptor relationship (SRR) of arriving air masses is evident in Fig. 3 (specifically within the 8–13 km layer shown in Figs. 3d and 3e), which influences both low and high O_3 over WNA. While this 8–13 km layer overlaps with the UTLS region shown in Figures 3 and 4 of the main manuscript, this altitude range also coincides with commercial aircraft cruise altitudes and frequent lightning activity. Consequently, it is difficult to attribute the increasing trend of SRR in the 8–13 km layer solely to a stratosphere-troposphere exchange (STE) signal.

To clarify these points for readers, we have incorporated the following discussion into the revised manuscript:

"Although the 8–13 km altitude range corresponds to the UTLS region and influences stratosphere-troposphere exchange (STE), it also intersects commercial flight corridors and regions of frequent lightning activity. Therefore, the upward SRR trend in the 8–13 km layer cannot be uniquely attributed to STE. Furthermore, we intentionally excluded extreme O_3 events from our primary analysis to mitigate STE influence. Investigating whether this increasing SRR trend points to a potential rise in STE activity requires further study and is beyond the scope of this work."

R2-8: Data availability

The OMI total ozone data including discussion of data quality are available from https://ozoneaq.gsfc.nasa.gov/.

I believe the dataset referenced here is Ziemke's tropospheric ozone product rather than total ozone. Since this satellite dataset is not directly used, I do not think it is necessary to provide the data source here.

A2-8: Thank you for pointing this out, and we apologize for the confusion. While we initially analyzed OMI total ozone as shown in Fig. R1. As mentioned in the response letter to Reviewer 1, we later decided to omit these data from the original manuscript because:

1) The OMI/MLS O_3 and NO_2 trends show patterns consistent with TCR-2 and GMI over Western North America, including consistent increasing trends.

2) TCR-2 already assimilates OMI satellite observations, so including the raw satellite data did not provide substantial new insights.

Because these results align with our findings and support the datasets used in this study, we have removed this statement from the revised manuscript.

References

Li, P., Yang, Y., Wang, H., Li, S., Li, K., Wang, P., Li, B., and Liao, H.: Source attribution of near-surface ozone trends in the United States during 1995–2019, Atmos. Chem. Phys., 23, 5403–5417, https://doi.org/10.5194/acp-23-5403-2023, 2023.

Ansari, T., Nalam, A., Lupaşcu, A., Hinz, C., Grasse, S., and Butler, T.: Explaining trends and changing seasonal cycles of surface ozone in North America and Europe over the 2000–2018 period: a global modelling study with NO_x and VOC tagging, Atmos. Chem. Phys., 25, 16833–16876, https://doi.org/10.5194/acp-25-16833-2025, 2025.

Ryoo, J.-M., Johnson, M. S., Iraci, L. T., Yates, E. L., and Gore, W.: Investigating sources of ozone over California using AJAX airborne measurements and models: Assessing the contribution from long-range transport, Atmospheric Environment, Volume 155, 2017, 53-67, https://doi.org/10.1016/j.atmosenv.2017.02.008, 2017

Citation: https://doi.org/10.5194/egusphere-2026-1157-RC2