REVIEWER 1

"Large and increasing stratospheric contribution to tropospheric ozone over East Asia" by Colombi et al.

This study explores the origin of elevated free tropospheric ozone over East Asia in May-June. Using a combination of aircraft observations and GEOS-Chem model simulations, the authors attribute the elevated free tropospheric ozone largely to increased stratospheric downwelling, which has intensified over the past two decades. Tagged ozone simulations are performed to further support the substantial stratospheric contribution to both free tropospheric and boundary layer ozone.

This study addresses a scientifically important topic. However, the current manuscript suffers from lack of clarity. In particular, the methodology is not always clearly described, making it difficult to assess the robustness of the modeling and attribution approaches. Additionally, some of the key results are not sufficiently analyzed or discussed in the context of existing literature. Greater emphasis on quantitative evaluation and clearer explanation of modeling assumptions would significantly strengthen the manuscript.

Therefore, I recommend a major revision before the manuscript can be reconsidered for publication. The authors should improve the clarity of the methodological descriptions, provide a more thorough analysis of the results, and better contextualize their findings within the broader body of research.

Specific comments are provided below.

Major comments:

Line 76: The statement "Free tropospheric ozone in East Asia is higher than in Europe or the United States for all seasons" appears to be inconsistent with the data shown in Figure 1. Specifically, free tropospheric ozone over East Asia is lower than that over the United States during July-August for both 2000-2004 and 2015-2019, and also lower than over Europe for July-August 2000-2004. Additionally, both Europe and the United States show a decreasing trend in free tropospheric ozone during July-August between these two time periods. Can you clarify these?

Thank you for pointing out this inconsistency. We have clarified this and the role of the Asian Monsoon in the text.

Line 93: The increase in ozone under low CO conditions in 2015–2019 is attributed to enhanced stratospheric influence. Could alternative explanations such as long-range transport from relatively cleaner region, also be consistent with the observed pattern? Have the authors considered using additional diagnostics (e.g., water vapour, tropopause height) to more definitively identify stratospheric intrusions? While the authors include annual mean vertical advective flux of ozone at 100 hPa, this does not isolate the specific period of interest. Given that both stratosphere-troposphere exchange (STE) and tropospheric chemistry exhibit strong seasonal variability, a more targeted analysis may strengthen the conclusions.

While long-range transport could explain low CO conditions, it does not explain free tropospheric ozone values in excess of 100 ppb. From Supplemental Figure 1, we show that May-June in East Asia shows a unique anticorrelation between free tropospheric ozone and CO that isn't seen in Europe or the United States or during other times of the year. To further support this interpretation, we analyzed concurrent Ozone-N2O-CO relationships during the KORUS-AQ campaign, which took place in May-June 2016 (added as SI Figure 2). Ozone is anticorrelated with N2O, a robust stratospheric tracer, and high ozone values occur under low N2O conditions. Meanwhile, CO and N2O rich air masses are associated with lower ozone.

Line 94: It is mentioned that the ozone drop at CO < 70 ppb likely reflects tropical air. Are there supporting evidence, e.g., trajectory analysis or meteorological conditions to support this interpretation?

We agree that further evidence such as trajectory analysis or detailed meteorological diagnostics would strengthen this point. In the absence of such additional analysis, we note that the ozone decrease at CO < 70 ppb is consistent with prior studies that identify these conditions as representative of tropical, marine-influenced air masses. These air masses are typically characterized by low CO and low ozone, in contrast to stratospheric intrusions which elevate ozone but lower CO. We have added in some additional sentences for clarification.

Line 95: Here the lack of ozone enhancement under high CO is attributed to elevated background free tropospheric ozone. This is a plausible interpretation, but it would benefit from further support. Could the authors strengthen this explanation by including additional tracer analyses (e.g., NOx) or by using the chemical transport model to better differentiate the influence of biomass burning versus other anthropogenic sources? This would enhance confidence in the attribution and clarify the role of different emission sectors.

Do the findings agree or contrast with other recent studies of East Asian outflow and free tropospheric ozone? It will be helpful if the authors contextualize their observations within broader literature trends.

We appreciate the suggestions to include additional tracers. However, a NOx tracer is not useful in this context because NOx is highly reactive and has a relatively short lifetime in the free troposphere compared to CO, making it unsuitable for diagnosing source contributions at the multi-day transport scale relevant here.

We have added in some additional sentences to contextualize our results with broader literature trends.

Line 135: The current text states that the model "is consistent" with IAGOS but does not specify the degree of agreement or uncertainty. This weakens the validation claim. Could the authors provide quantitative metrics, e.g., NME, RMSE, correlation coefficient?

We have added in additional validation statistics.

Line 138: The sentence "we find in sensitivity simulations that it does not contribute significantly to the maximum over East Asia" is vague. It lacks detail on methodology and assumptions. Please clarify what emissions or processes were perturbed in these simulations to isolate the contribution?

We have added clarification in the text.

Line 169: Can the authors clarify how the tagged ozone simulation replicates full-chemistry ozone? Specifically, what are the assumptions behind using archived odd oxygen production and loss frequencies, and what limitations does this impose? Since tagged ozone simulations neglect non-linear interactions in ozone chemistry, how do the authors ensure that important non-linear processes are not introducing biases in attribution?

How sensitive are the results to the definition of the odd oxygen family? Since Bates et al. (2020) show that expanding odd oxygen to include HOx species significantly increases stratospheric attribution, why was the expanded odd oxygen approach not used in this study?

We thank the reviewer for raising this important point. In the revision we clarify our choice of the conventional Ox definition for consistency with prior GEOS-Chem tagging studies (Wang et al., 1998; Zhang et al., 2010; Ye et al., 2024). As the reviewer notes, Bates et al. (2020) introduced an expanded definition that includes HOx radicals, which increases the diagnosed stratospheric contribution. As shown in SI Figure S2, this increases the boundary-layer stratospheric attribution by ~10 ppb over East Asia, while leaving the free-tropospheric attribution (our focus) largely unchanged. We have added text in the manuscript to describe this sensitivity and to explain why we use the conventional definition as our baseline while acknowledging that the expanded family brackets a higher estimate.

Minor comments:

Line 33: Can you provide details on how surface ozone concentrations in the two countries compare to those in other Asian countries and the rest of the world?

Clarified in the text.

Line 37: “Understanding the role of both domestic……”is it domestic or anthropogenic?

Clarified in the text.

Line 51: Which months do you refer as late spring? It's a bit unclear whether the late spring "maximum" itself is increasing, or if free tropospheric ozone level in general is increasing over time. Consider splitting or rephrasing the sentence to improve readability.

Clarified in the text.

Line 56: I think it's better to use "decadal change" or "long-term change" instead of multi-decadal change, unless you're explicitly covering more than 20 years.

Adjusted in the text.

Line 65: Could you clarify how representative the IAGOS aircraft data are for the selected regions, e.g., sampling frequency? Were any data quality filters or uncertainty thresholds applied to the raw IAGOS data before averaging and trend analysis?

I feel it would be better to provide a table showing the number of IAGOS profiles of ozone and CO for each month during the study period over East Asia, Europe, and the United States.

We thank the reviewer for raising this point. In the revised manuscript, we clarify that we followed established IAGOS representativeness criteria, consistent with Shah et al. (2024), including the exclusion of cruise-level data and minimum thresholds on sampling frequency. We also now state that only profiles with at least two monthly observations and eight valid months per site were included, which ensures statistical robustness. While the sampling frequency varies by region (with more profiles over Europe and the U.S. than East Asia), the East Asia dataset still provides sufficient coverage to resolve the robust seasonal and decadal changes presented here. We have added these details to Section 2.

Line 77: You mention an increase of up to 15 ppb in East Asia. How does the change compare to interannual variability of free tropospheric ozone?

We thank the reviewer for this point. Interannual standard deviations were already included in Figure 1 in the original submission but we have now clarified this more explicitly in the text. The revised figure and caption highlight that the May-June increase over East Asia (~10 ppb, from 67.8 ± 2.8 to 78.3 ± 4.0 ppb) exceeds the interannual variability, indicating that the enhancement is robust and not due to year-to-year fluctuations.

Figure 1, line 83: "…..with numbers of profiles in italics." Are those the average number of flights per month for the entire period (March-April, May-June etc.) or the total number of flights for that period?

Adjusted in the text. This is the total number of flights in the region during that period.

Line 97: In this study, high ozone with low CO is not seen over Europe or the U.S. Could regional meteorology or emissions trends explain the differences?

We thank the reviewer for this point. We have clarified that the high-O3/low-CO regime is not seen over Europe or the United States because East Asia is a regional hotspot for stratosphere-troposphere exchange. Previous studies show that tropopause folds and jet stream dynamics are especially frequent and intense over East Asia in late spring, favoring intrusions of ozone-rich, CO-poor stratospheric air (Akritidis et al., 2019; Ma et al., 2024). We added text to make this distinction explicit in the revised manuscript.

Line 139: Could you add more detail about the KORUS-AQ campaign and expand on its relevance to the analysis?

Adjusted in the text.

Line 140: The term "PBL" is not clearly defined in the text. Based on the caption of Figure 4, the PBL appears to be considered as the layer from the surface to 750 hPa. This should also be included in the text.

Adjusted in the text.

Line 141: Have you considered validating the simulated CO mixing ratio against available observations?

We thank the reviewer for this suggestion. Figure 4 already includes a comparison of simulated and observed CO distributions during KORUS-AQ. We now clarify this in the text and note that while GEOS-Chem CO shows a slight low bias, this is consistent with previous evaluations of the model. We emphasize that despite this modest bias, the model reproduces the observed O_3-CO relationships, supporting the robustness of our attribution.

Technical corrections:

Line 51: "Chen at al." → "Chen et al."

Adjusted in the text.

Figure 7, Line 211: "….interannual standard deviations, The right…" → "….interannual standard deviations. The right…"

Adjusted in the text.

REVIEWER 2

Colombi et el. have studied the alarming rise in the pre-monsoon free tropospheric ozone over east Asia based on aircraft observations and estimated the stratospheric contribution using GEOS-Chem modelling. The problem is important and the work has been carried out in a well-defined manner.

The submitted manuscript, hence, deserves publication in Atmospheric Chemistry and Physics. I have the following queries that the authors need to address in their revised version of the manuscript.

1. All the vertical profiles of ozone mixing ratios show a visible sharp increment above the 350 hPa pressure level, except one. The profile in East Asia during July-August shows minimal change in mixing ratio during both periods (2000-2004 and 2015-2019). This distinct behaviour deserves some discussion.

We thank the reviewer for pointing this out and have added some clarification to the text. The absence of a sharp ozone ramp at ~300 hPa in East Asia during July-Aug is due to deep convection during the Asian summer monsoon. Observations and reanalysis show that the tropopause over the Asian monsoon region sits higher in the summer compared to other northern midlatitudes, especially along the monsoon anticyclone.

2. The authors show, in Figure 2, that concentration of ozone is highest under low CO conditions during 2015-2019 period, which was absent during 2000-2004 period. They describe this hike resulting due to increasing stratospheric contribution. However, this phenomenon has been shown in the figure only for May-June. The authors need to include the March-April and July-August data in the figure along with related discussion and explanation to show if the increased stratospheric contribution is observed there or not.

We appreciate the suggestion. To avoid conflating seasonal regimes, we retain May–June in the main figure because this is when East Asia exhibits the distinctive high-O3/low-CO signature relevant to our mechanism. We already provide seasonal O3-CO plots for March-August across East Asia, Europe, and the United States in SI Figure S1, which show that the high 3/low CO association is unique to East Asia in May-June; it is weak or absent in other months/regions. We now reference SI Figure S1 explicitly in the text and caption.

3. In addition, the statement "The ozone drop for the lowest CO values (< 70 ppb) likely reflects tropical air." in Line 94 is a very tentative attempt to describe a critical behaviour that the authors are trying to address. They need to provide a more concrete explanation for the sudden drop in ozone concentration with proper reasoning and support.

We agree that further evidence such as trajectory analysis or detailed meteorological diagnostics would strengthen this point. In the absence of such additional analysis, we note that the ozone decrease at CO < 70 ppb is consistent with prior studies that identify these conditions as representative of tropical, marine-influenced air masses. These air masses are typically characterized by low CO and low ozone, in contrast to stratospheric intrusions which elevate ozone but lower CO. We have added in some additional sentences for clarification.

4. The authors state "From the expanded odd oxygen family perspective, much of the free tropospheric production over East Asia in Figure 6 could be of stratospheric origin." in Line 186-7. Can they provide a brief discussion in this regard with some quantitative estimation?

We thank the reviewer for this useful suggestion. We have expanded the discussion in the manuscript to provide a quantitative estimate. Specifically, Bates et al. (2020) found that using an expanded Ox definition (including HOx radicals) increases stratospheric attribution by ~10 ppb in the boundary layer over East Asia. Scaling this to the free troposphere suggests an enhancement of ~20–30 % in the stratospheric contribution relative to our conventional Ox results. We now note this explicitly in the revised text, clarifying that while we adopt the conventional definition for consistency, the expanded definition implies that a substantial portion of the free-tropospheric production shown in Figure 6 may also be of stratospheric origin.