Reviewer 2:

Reviewer #2 (Comments to Author (shown to authors)):

The manuscript by Linda Ort and other co-authors provides a study combined in situ aircraft observations of O_3 and CO from 12 global research campaigns conducted between 2012 and 2024, covering different seasons and latitudes from the boundary layer up to the lower stratosphere, with simulations from the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model. Two simulations were performed: a reference run including all standard emissions, and a sensitivity run with lightning NO_x emissions turned off. The authors demonstrated that observations and model both show a pronounced enhancement of the O_3-CO ratio in the northern subtropics, extending nearly to the boundary layer. They conclude that tropospheric photochemistry via tropical lightning NO_x in the upper tropical troposphere drives high O_3-CO ratios in the subtropics, which is traditionally attributed to stratosphere-troposphere exchange (STE) near subtropical jets.

The scientific significance of this manuscript is absolutely given, as it is important to clear understanding the effects of transport and photochemistry via Hadley circulation in the upper troposphere of the tropics to the subtropics on variations of O_3 and CO. This dataset is very valuable, especially given the scarcity of such measurements with high vertical resolution. I have several concerns regarding the integration of observations and simulations. Below, I outline the main issues and offer suggestions for improvement.

We thank Reviewer #2 for the time reviewing our manuscript and for underlining the significance of our results and the thoughtful questions. Here blow the Reviewer's comments are repeated with our answer.

Major comments:

1) Multiple instruments measured high-resolution CO (e.g., UMAQS, QCLS, TRISTAR, ATTILA) and O_3 (e.g., FAIRO UV absorption, chemiluminescence) and O_3 during these campaigns. Most of them implemented in one month spanning 12 years. The authors averaged all observations to 60 s and gridded at 1 km vertical × 1.875° latitudinal resolution to evaluate model performance. Given the relatively rapid mixing timescales within and between tropospheric hemispheres (and between the troposphere and stratosphere), simple averages of measurements in different campaigns likely biases the seasonal signal. I recommend comparing the model against co-located, time-matched observations to assess its ability to reproduce monthly and seasonal variability.

We understand the reviewer's concerns regarding the averaging of different instrumentation, campaigns and seasons. To avoid bias introduced by the selected campaigns, we have tested the representativeness of the extracted flight tracks against the full climatology of the model. This has been shown in Figure S1 (now S2) in the Supplement, where "ft mod" represents the 1-hour-resolved co-located and time-matched modeled data, following a nearest point algorithm, and "mod" the full modeled climatology. Using a higher resolution (60 s) or a direct extraction through interpolation of the model would indeed result in more detailed comparisons between the model and the observations. However, as we are interested in the climatological implication, we believe it is more instructive to compare the climatological data to the observations.

Furthermore, this study mainly focuses on an annual and large-scale transport process, which makes us confident the comparison suffices for our analysis.

However, to convince Reviewer #2, that the bias obtaining in averaging 12 years, various instruments, and 12 different campaigns, is negligible, we have added Figure AC2, showing the correlation between the hourly-resolved O_3-CO ratio of the observations against the O_3-CO ratio extracted from the model along the flight tracks, the same data set used for "ft mod", color-coded by the campaigns, and size-adjusted according to the observation altitude. The 2*RMSE shaded area and an R^2 of 0.41 show that most of the data points are within the confidence intervals, and no campaign, year, nor instrument is showing a significant bias. Nevertheless, the spread increases towards larger values of the O_3-CO ratio and at higher altitudes. This shows the underestimation of O_3 towards the tropopause, which is likely due to the grid resolution of the model, and which we are taking into account in the discussion and clarified in Line 142.

More detailed evaluations of most of the campaigns and the EMAC model, also in higher resolution, have been published previously, which we are addressing and citing in Section 2.2.

Line 142: "O_3 overestimation by the model is attributed to an overestimation of transport from the stratosphere, related to the limited horizontal grid resolution of the model (1.875° x 1.875°) (Lelieveld et al., 2018). However, by reducing LNOx emissions associated with deep convection, photochemical O_3 production declines in the upper troposphere."

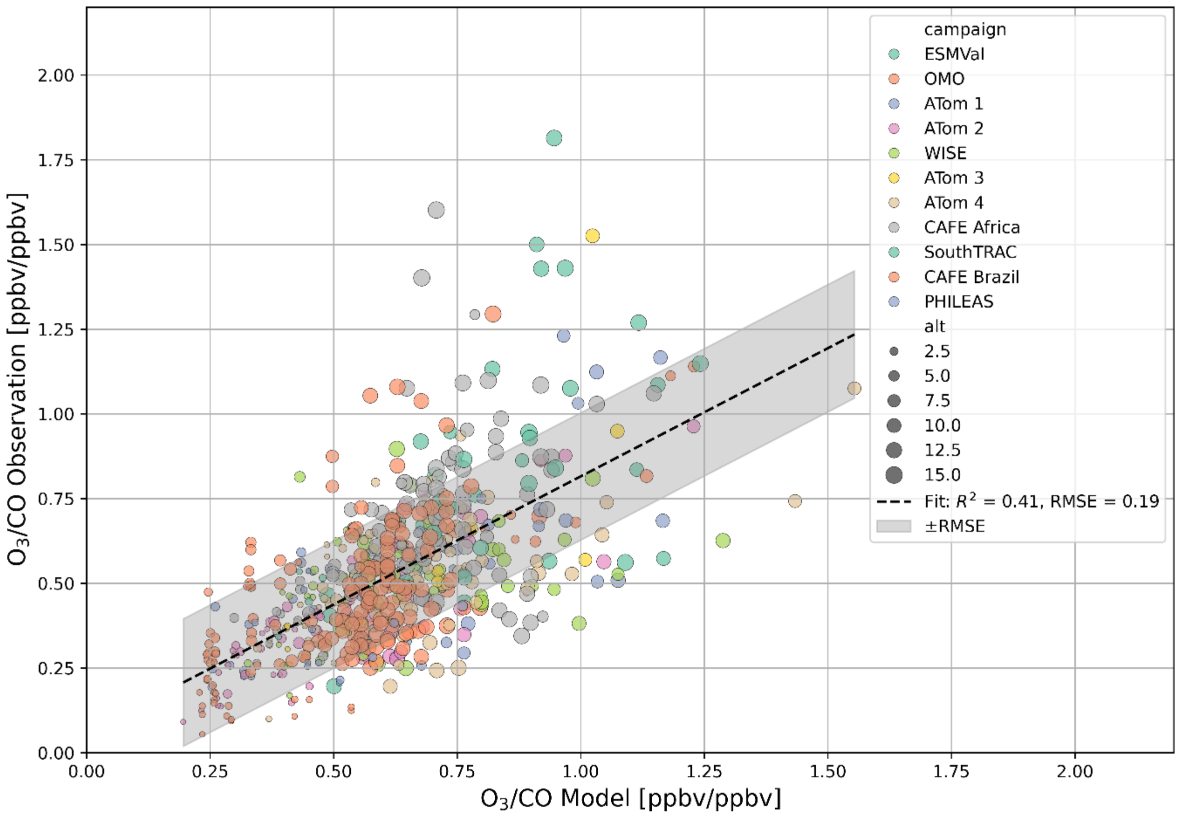

Figure AC1: Comparison of observed and modeled O_3-CO ratio from the 1-hour resolved comparisons. The size of the dots visualizes the measurement height and the color coding indicates the respective campaign. The black line shows the non-weighted linear regression and the shaded area the root mean squared error (2*RMSE), representing the confidence area around the linear fit.

2) The current analysis lacks a direct comparison of observed and simulated high-resolution vertical profiles. Such a comparison would be invaluable for diagnosing the seasonal behavior of STE on a monthly timescale and for validating the vertical transport processes in the model.

That is an interesting point. High-resolution monthly vertical profiles of observations could tell us more about the seasonal behavior of STE, and further validate and improve modeled STE. However, such high-resolution data sets, vertically scanning the troposphere with no immediate influence close to source regions (e.g., airports, cities), from airborne in-situ measurements are not yet available on a monthly timescale. Even the collection of the 12 different aircraft campaigns used in this study does not cover enough data for monthly comparisons. Hence, we decided to focus on an annual average, as we can at least assure no seasonal or campaign bias, which we have shown in the direct comparison of the modeled flight-track extracted data with the climatology shown in the supplement in Figure S2.

However, the ATom missions are closest to a high-resolution data set, as those flights covered the same tracks over four seasons. We have plotted the vertical profiles of O_3 within the northern subtropics (23.3° - 40°N) of the four ATom campaigns together with modeled O_3 and the stratospheric O_3S tracer from the EMAC model in Figure AC3. Please note that each ATom campaign typically lasts only two months. For the vertical profiles of modeled data, we select the same latitude range and months according to the observations, but from the full climatology data set of the model.

Seasonal differences in STE are mostly dependent on the position of the tropopause, being highest in summer (JUL-AUG) and lowest in winter (JAN-FEB). Those seasonal aspects are simulated by the model. However, the general overestimation of O_3, which we have discussed and addressed in the manuscript in Sections 2.2 and 3.2, occur in those vertical profiles as well.

Clearly, a detailed investigation in the behavior of STE would be really interesting, but, unfortunately, goes beyond the scope of this work, as more global observations are needed and a more detailed meteorological analysis would be needed. This study aims to highlight and address the importance of tropospheric chemistry on the global budget of O_3 and CO, which is, as shown, not easily distinguishable from stratospheric influence in the subtropics.

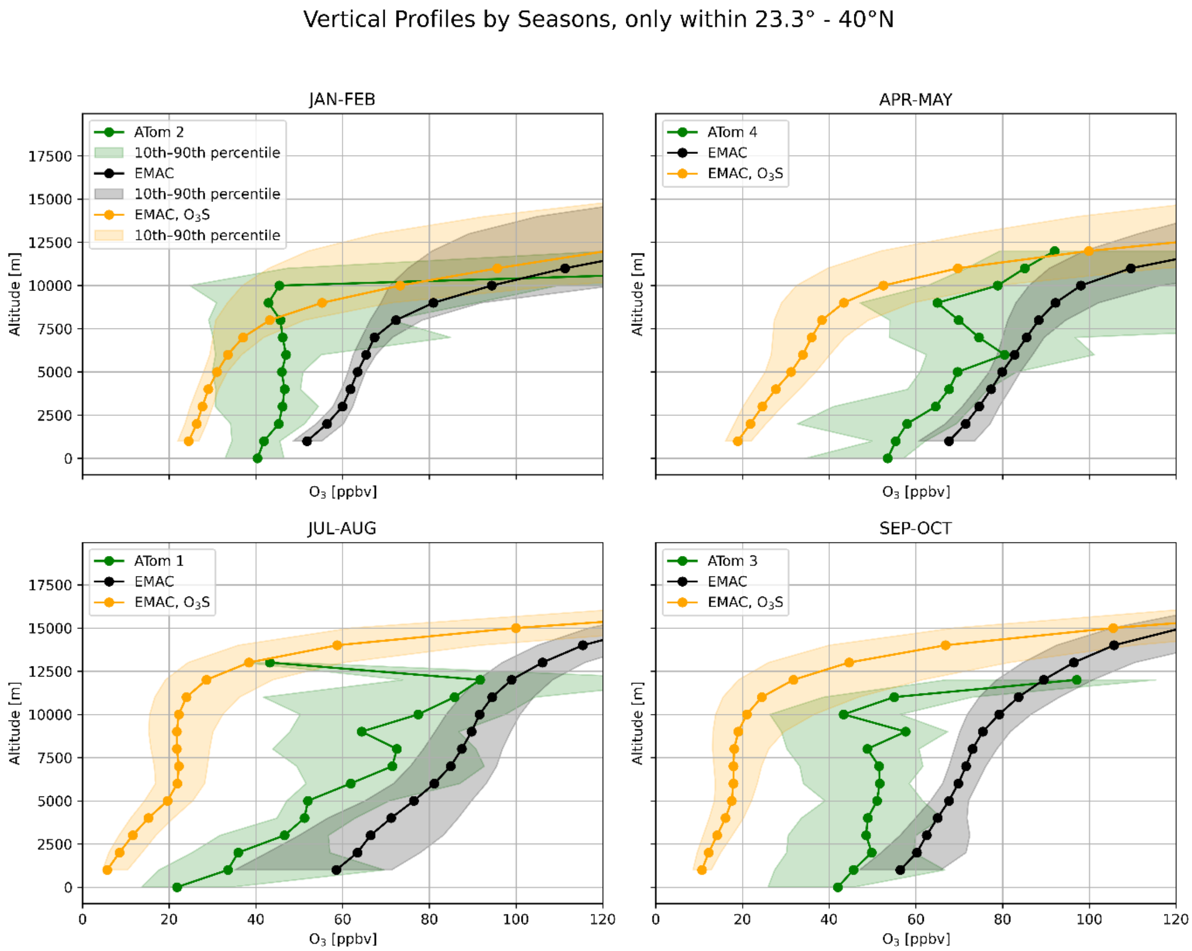

Figure AC2: Vertical profiles of O_3 of the four ATom missions (green), the EMAC modeled O_3 of the full climatology (black), and the O3S tracer of the EMAC model, representing O_3 entering the troposphere from the stratosphere (orange) within the northern subtropical latitude band, spanning from 23.3°N to 40°N. The dots represent the medians and the shaded areas the 10^{th} to 90^{th} percentiles. Each tracer was averaged over the two months during which the ATom mission was carried out.

3) the authors used simulations from ECHAM5/MESSy Atmospheric Chemistry (EMAC) model, why not ECHAM6? Given that ECHAM6 is available, with higher spatial resolution and improved representation of seasonal and intra-seasonal transport, I suggest justifying the choice of ECHAM5 or, if feasible, repeating key simulations with ECHAM6 to determine whether the results are robust to model version.

Based on the ECHAM6 manual (Giorgetta, Marco A., et al. "The atmospheric general circulation model ECHAM6-model description." (2013).), " Significant differences between ECHAM5 and ECHAM6 concern the land processes, the radiation schemes, the computation of the surface albedo, and the triggering condition for convection. [...] The spectral transform dynamical core and the flux form semi-Lagrangian transport scheme remain essentially unchanged." Apart from the land surface adjustments (addressed in recent work introducing the JSBACH land model), e.g., radiation, boundary layer processes have been updated in our version of EMAC, in which only the spectral dynamical core of ECHAM5 has been kept from the original code.

Therefore, the updates in ECHAM6 are not expected to directly affect our results. Importantly, no significant resolution improvements are available, as both models are spectral models, without changes in the solution of the basic equation of momentum.

Furthermore, the EMAC model has been nudged to the ERA5 data (Jeuken et al., 1996; Hersbach et al., 2020), and therefore the tracer transport follows that in the ERA5 dataset. Any impacts of changes in model version on tracer transport would be strongly moderated by the nudging procedure.

On the other hand, the representation of convection could directly influence our results. Nevertheless, the EMAC model version of ECHAM5 has been further developed, and much additional work has been performed over the years on this topic, with a detailed analysis and evaluation of convection (Tost et al., 2006, 2009, 2010), not available in the ECHAM6 model.

Therefore, we do not expect that a simulation with ECHAM6 will provide additional scientific insight into the topic. In future (currently in development), a simulation using a GCM coded with different dynamical core routines (such as ICON) would be very helpful and provide insight into the robustness of our results. The implementation of the MESSy interface into ICON is ongoing work, and this tool is not yet available.

Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., et al.: The ERA5 global reanalysis, Quarterly Journal of the Royal Meteorological Society, 146, 1999–2049, 2020.

Jeuken, A., Siegmund, P., Heijboer, L., Feichter, J., and Bengtsson, L.: On the potential of assimilating meteorological analyses in a global climate model for the purpose of model validation, Journal of Geophysical Research: Atmospheres, 101, 16 939–16 950, https://doi.org/10.1029/96JD01218, 1996.

Tost, H., P. Jöckel, and J. Lelieveld. "Influence of different convection parameterisations in a GCM." Atmospheric Chemistry and Physics 6.12 (2006): 5475-5493.

Tost, Holger, et al. "Uncertainties in atmospheric chemistry modelling due to convection parameterisations and subsequent scavenging." Atmospheric Chemistry and Physics 10.4 (2010): 1931-1951.

Tost, H., et al. "Convection parameterisation influences on trace species profiles." EGU General Assembly Conference Abstracts. 2009.

4) I suggest the authors focusing on understanding the monthly variations and mechanism of enhancement of O_3-CO at tropospheric subtropical latitudes, because that is very important for accurate projections.

We highly agree with Reviewer #2 that understanding the monthly variations and mechanism of the enhancement of the O3-CO ratio in the tropospheric subtropical latitudes is of great interest. With the section 3.3.4 “Seasonal variability” we suggest, that seasonal variation is coming from other atmospheric NOx sources than from lightning NOx and/or mixing from the stratosphere. Our study focused on the importance of tropical lightning NOx and its influences on photochemistry, transported over the Hadley circulation towards, a. o., the subtropics, which has an important influence on trace gas global distributions with a rather constant seasonality. Therefore, we outlined the need of further investigations on other mechanisms influencing O3 production and CO removal in the subtropics in the outlook (Line 523), and think that thermal depletion of PAN might be another NOx source to consider, which we mentioned in Line 491. But this needs more investigation and is beyond the scope of this work.

Lines 523-524: “Furthermore, other tropospheric sources of NOx seem to play an important role in conditioning the troposphere as well, which needs further investigation.”

Lines 491-493: “Possible transport of PAN via the Ferrell circulation from the mid-latitudes into the subtropics and its thermal depletion in the downward branch is another potential source of NOx.”

5) L234: Is “pols” a typo? Should be “poles”?

We have corrected this typo in the manuscript.