At first we want to thank the reviewer for the very helpful comments to improve the manuscript.

The manuscript "Impact of South American biomass burning emissions on elevated South Atlantic upper-tropospheric ozone" by Smoydzin et al. presents an analysis of measurements taken during two HALO flights in 2019. One of the flights showed large O3 and CO values, likely influenced by biomass burning. Using trajectory analyses and satellite data, the authors further interpret the measurements. I think the manuscript generally fits within the scope of ACP. In its current state, however, major revisions are necessary before it can be accepted for publication. Alternatively, I suggest converting the manuscript into a measurement report.

Main comments

(1)
The data analysis is well done and the two flights constitute an interesting case study. My main concern, however, is that the manuscript does not yet meet the definition of a research article in the journal. According to the ACP guidelines, research articles must include substantial advances and general implications for the scientific understanding of atmospheric chemistry and physics. In my opinion, the manuscript currently does not fulfil these criteria, especially because the authors provide a lot of references to previous analyses with similar findings (l52ff).

The scope of ACP states: "Articles should have important and clearly argued implications for our understanding of the state and behaviour of the atmosphere and climate or present substantial new insights into the atmosphere's role in other parts of the Earth system"
We combine novel aircraft measurements (yet unpublished) with latest state of the art Lagrangian and Eulerian modeling to disentangle the processes leading to upper tropospheric ozone- production based on the observation of continental-wide pollution enhancements in the UT. As stated by reviewer 1, 'this is probably the best and most comprehensive approach based on novel methods and also addressing a new aspect regarding the impact of biomass burning versus biogenic versus lightning NOx conditions.' Our results highlight the role of the effects of these processes for UT ozone and therefore enhances our understanding of state and behaviour of the atmosphere - even though just showing one example.
Most previous publications dealing with the South Atlantic O_3-maximum were from times, when the implications of VOC chemistry on O_3 formation, in particular in relation with convective uplifting were not known, yet. None of the other publications presents neither a chemical analysis of the O_3 formation processes nor any kind of chemical modeling to explain which processes control to which degree upper tropospheric O_3 formation.
We do exactly that.
Reviewer 2 states very correctly, that elevated upper tropospheric O_3 mixing ratios could as well be the result of solely biogenic VOC and CO emissions. We briefly discussed this in the first version of our manuscript (scenario $FLASH_{BL}$) and following the other reviewer, we extended this section considerably.
The discussion of biogenic versus biomass burning emissions and their impact on upper tropospheric O_3 chemistry over the South American continent and South Atlantic ocean has not been part of any other study before.

The observations presented here for the first time are unique for various reasons:
() Other observations over South America are more than 2 decades old and do not provide observations of a comprehensive set of chemical trace species like for the SOUTHTRAC campaign. In-situ measurements include: O_3, NO, NO_y, N_2O, C_2H_6, HCOOH, CH_3OH, C_2H_4, PAN, CO, as well as remote sensing observations of various species.
() The work of e.g. [Jenkins et al., 2021] is solely based on the analysis of 20 ozonesondes. Sonde data are spacially and temporally limited and do not include measurements of other chemical trace species.

Other studies only present O_3 columns retrieved from satellite measurements. Hypothesis made by e.g [Jenkins and Ryu, 2004] are very general (*it is likely that winds in combination with lightning from West Africa, Central Africa and South America are responsible for enriched upper tropospheric* O_3), whereas we try to disentangle the role of different precursors to ozone formation.
() The observations taken onboard the HALO aircraft are unique as 2 flights were performed at exactly the same altitude, longitude and latitude over a distance of hundreds of kilometer. This is a unique data set particularly given the consistency and combination of measurement data allowing for a comparison between the flights.

() Many publications about biomass burning in general exist, but as stated above, our study adds new aspects based on state of the art data, model based analysis and a new metric. Especially therefore we have to cite related work in the introduction and to compare own work with other studies.

() The novel observations however, are just the motivation for this study. The scientific explanation of the observations and thus our results are 100% based on a novel combination of combined model-based methods in addition to a metric, which was first published in 2023 (Nussbaumer et al.,). The combined methods include a set of four different numerical models on different scales (EMAC=global chemistry-climate model, ERA5-weather forecast and re-analysis model, MPTRAC - lagrangian trajectory model, CAABA - chemical box model) in addition with MOPITT CO satellite data. Based on the novel metrics by Nussbaumer et al. 2023, we present a detailed analysis of O_3 production regimes differentiating lightning and biomass burning impact. Finally, we calculate the radiative impact of the biomass burning induced observed O_3 enhancement.

We do consider this as quite a lot of new content leading to novel scientific result namely the importance of biomass burning generated VOCs for O_3 production in lightning generating convective tropical regimes. The latter being not sufficient to explain the observed ozone production.

The authors attempt to contextualise the measurements with CO satellite data, but CO alone does not fully describe the situation. I wonder: Does ozone show similar enhancements in this region each year and is it caused by biomass burning? Insights from either satellite data (which may be complex) or model results would help.
The intention of this manuscript is not a climatological study but we present 2 case studies showing a continental-scale impact. It is also common research method - case studies are published in this journal continuously - by the way also for regions for which numerous case studies already exist (e.g. eastern or western US). As mentioned above, we already include data from 4 different numerical models + CO satellite data + radiation calculation + observations. Including a further dataset would go far beyond the scope of this paper.
Note, that results from a model simulation neglecting the biomass burning have been included in the revised version of the manuscript to further strengthen the importance of biomass burning emissions.

To satisfy the journal's criteria the authors would need to place their results into a broader context, e.g. by addressing questions such as:

(1) How typical/atypical were the emissions during that period?
(2) How common are such high ozone values caused by biomass burning in this region? (Based on the above)
(3) What is the radiative-budget impact of these events?

(1) Please have a look at Fig. 6 showing MOPITT CO mixing ratios. CO is a very strong

and commonly used marker for biomass burning emissions. The annual cycle of tropospheric CO level follows the annual cycle of biomass burning emissions which are strongest in the burning season (SEP - OCT) and are lower in the transition seasons and the wet season (DEC-FEB). As mentioned in section 3.4, line 322-327, CO mixing ratios for the year 2019 for Sep and Oct (the 2 months of the flight) are exactly on (OCT) or near (SEP) the regression line for the 20 years of available MOPITT-CO data. Therefore it can be concluded that atmospheric conditions prevailing during the research flights can be considered as typical.
(2) There is no existing dataset giving a vertically and horizontally resolved climatology of O_3 level in the atmosphere, which allows to differentiate between the troposphere and stratosphere. Based on IASI satellite data, [Tsivlidou et al., 2023] show that upper tropospheric O_3 mixing ratios over the South Atlantic are significantly higher in OCT than in other seasons. In addition, other observations cited by us in the introduction section also report elevated upper tropospheric O_3 level over the South Atlantic.
(3) We discussed (abstract and section 3.5), the radiative impact of the enhanced O_3 level in the UT and compare this result with the work by Rap et al. (2015) (section 3.5) and the shortwave direct aerosol radiative effect (l.360-362). In the revised version we extended this discussion and show the combined shortwave and longwave effect locally at the tropopause and at the top of the atmosphere.

The answers to these questions are particularly important because the authors calculate an effect of ozone on the radiation budget. The 50mWm-2 figure they report is not very useful without a larger picture.
As stated above, IASI satellite observations indicate substantial ozone enhancements also over the Atlantic [Tsivlidou et al., 2023] indicating transport of produced ozone. Since our data are limited we don't want to speculate on the vertical column extent of the ozone enhancement and decided to keep the figure as it is.

I understand that these analyses will require substantial additional work. Therefore, publishing the manuscript as a measurement report might also be an option. In that case, only the minor comments below need to be addressed.
Please refer to the answer we have given in (1)

The second main comment concerns the radiation calculations. Many details of the model and the exact methodology are unclear. Please specify them. The text and figures are also difficult to follow. As already mentioned by the reviewer, it is also not clear where the numbers in the abstract come from.
We slightly extended the model description referring to a description of the radiation calculations. The vertical profiles of the radiatively active compounds have been taken from a global chemistry-climate model simulation using the EMAC model. Effects of aerosol particles and cloud effects have been ignored for these calculations, as the differences from the effects of O_3 are of higher relevance than the absolute numbers of the radiative fluxes.

Minor comments
Please check the usage of the word "scenario" with respect to the model runs. In my understanding you do not consider emission scenarios (which would refer to future developments) but rather different sensitivity/emission cases.
It is correct, that we don't apply future scenarios, but in our case it is obviously a present-day emission scenario. The usage of the term 'scenario' is not restricted to 'emission scenarios'. It is a common habit to refer to model simulations using different model setups (also for present-day conditions) as 'simulation scenarios'.

L279: The authors write that EMAC model data are on a 1×1deg grid, but on L158 they mention T42L90MA, which would be 2.8x2.8deg. Please clarify.
The emissions are provided on a 1x1 degree grid. For usage in EMAC they are regridded to the respective model resultions. As the boxmodel and the Lagrangian trajectories do not require such

a regridding the high native resolution of the emission data can be used for the CAABA simulations. The chemical initial conditions are taken from EMAC (originally on the T42 grid) and interpolated to the corresponding trajectory point.

L164: If you use RCP6.0 emissions, they are outdated and may not reflect the "real" emissions in 2019. How does this affect your results? Could anthropogenic CO/NOx emissions in the region of interest have increased strongly compared to RCP6.0? This is important because it would strongly influence your BB trajectories.
Anthropogenic emission over Amazonia are generally very small. Stronger anthropogenic emissions would hypothetically affect all trajectories which have BL contact - not only BB trajectories. However, we can consider it as state-of-the art knowledge that over Amazonia (rain forest region), biomass burning emissions and biogenic emissions exceed anthropogenic emissions by far. The potential increase of CO and NOx emissions compared to RCP6.0 can be considered minor compared to the strength of biogenic/ biomass burning emissions. In addition, it is to mention that all anthropogenic emission inventories underlay an uncertainty.

Figure 4: Please explain the colours in the bar fractions. Is the yellow the same as FLASH_noBL? If so, please use the same colour.
We checked the color codes of both corresponding figures. The colours for the bars are exactly the same as the colour code in the legend of Fig. 4a (red BB, bright yellow FLASH_noBL, dark yellow FLASH_BL....) and bars have also exactly the same colours as the marks in the line plot below the bars.

References

[Jenkins et al., 2021] Jenkins, G. S., de Castro, V., Cunha, B., Fontanez, I., and Holzworth, R. (2021). The evolution of the wave-one ozone maximum during the 2017 lasic field campaign at ascension island. *Journal of Geophysical Research: Atmospheres*, 126(10):e2020JD033972. e2020JD033972 2020JD033972.

[Jenkins and Ryu, 2004] Jenkins, G. S. and Ryu, J.-H. (2004). Space-borne observations link the tropical atlantic ozone maximum and paradox to lightning. *Atmospheric Chemistry and Physics*, 4(2):361–375.

[Tsivlidou et al., 2023] Tsivlidou, M., Sauvage, B., Bennouna, Y., Blot, R., Boulanger, D., Clark, H., Le Flochmoën, E., Nédélec, P., Thouret, V., Wolff, P., and Barret, B. (2023). Tropical tropospheric ozone and carbon monoxide distributions: characteristics, origins, and control factors, as seen by iagos and iasi. *Atmospheric Chemistry and Physics*, 23(21):14039–14063.