Review of "Impact of South American biomass burning emissions on elevated South Atlantic upper tropospheric ozone" by Linda Smoydzin, Vera Bense, Heiko Bozem, Philipp Joppe, Daniel Kunkel, Hans-Christoph Lachnitt, Holger Tost, Andreas Zahn, Helmut Ziereis, Martin Riese, and Peter Hoor"

In this manuscript, the authors examine the impact of South American biomass burning emissions on the increase in ozone levels in the upper troposphere over the South Atlantic. For this study, the authors utilize a comprehensive set of observational data and modelling. They combine airborne in situ observations from two flights of the 2019 SOUTHTRAC HALO mission, remote sensing satellite data, back trajectory analysis, chemical Lagrangian box modelling and the EMAC model.
The manuscript is logically organized and the research is in line with the overall subject areas of ACP. However, the authors need to be more precise in some cases (see further minor comments). In the current state of the Discussion and Conclusions section I am not convinced that the results can support statements such as in line 344: "...we can conclude that biomass burning emissions contribute predominantly to the enhanced upper tropospheric O3 level...".

I would recommend the manuscript to be published after the following points were addressed in a minor revision.

Comments regarding the conclusions of this study:

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

The authors show that the BB scenario produces more ozone then the FLASH_BL and FLASH_noBL cases. However, as most trajectories are FLASH cases, the main contribution still comes from the FLASH cases (Fig 4b). The authors rightfully argue that the influence of boundary layer may be underrepresented by the model (for BB and FLASH scenarios) and demonstrate that the ozone production is limited by VOC.
However, in the description of the sensitivity simulations (lines 286ff) the added amount of VOC is not quantified. This leaves the question open if the "predominant" contribution is actually coming from VOC emitted by biomass burning emissions, or if biogenic VOC which is abundant over the South American rainforest (see e.g.: Tripathi at al. 2025) and are uplifted from the BL during convective events associated with lightning could explain a significant amount of the VOCs needed in the model to match the observations.
Without further quantification the presented results together with the also not quantified uncertainties of the simulations can't support the conclusion stated in lines 12, 308, and 344, that biomass burning emissions are the "predominant" contributor to the observed ozone enhancement. In the current state, the results suggest that the predominant contribution is lightning NOx associated with VOCs uplifted from the BL, also effected by biomass burning emissions.

We added large parts of the explanation below to the manuscript to facilitate understanding our simulation experiments and the conclusions drawn from them.

(1) Experiments with additional VOCs in scenario FLASH$_{\mathrm{noBL}}$:
We assume that at the selected grid points (CAPE > 1000 = CAPE$_{\mathrm{high}}$), EMAC (ambient) VOC mixing ratios are a factor two higher then actually simulated by EMAC. Thus, depending on the

number of $\text{CAPE}_{\text{high}}$ events along the trajectories, VOC mixing ratios in the CAABA box are successively enhanced along each trajectory (by mixing with ambient (EMAC) air masses). Thus, the more $\text{CAPE}_{\text{high}}$ events, the larger is the difference in VOC mixing ratios between our base simulation and the simulation with enhanced VOCs and finally the difference in O_3 mixing ratios. This finally leads to VOC mixing ratios of up to an order of magnitude larger at the flight track for scenario noBL compared to the base simulation.
We also performed a test, increasing EMAC_{VOC} mixing ratios everywhere for scenarios FLASH_{BL}, $\text{FLASH}_{\text{noBL}}$ and REST. An enhancement factor of 1.2 leads to the formation of the observed O_3 level. However, this also leads to simulated VOC mixing ratios at the flight track of up to 100ppb which we consider to be unrealistic.

(2) Furthermore, we have observations at the flight track of PAN and several organic species (C_2H_6, HCOOH, CH_3OH and C_2H_4, [Johansson et al., 2022]) with witch we can evaluate the CAABA simulations. In our base simulation (and consequently also in the simulation with enhanced VOCs), we have a good agreement in the temporal evolution of these species along the flight track but mixing ratios of C_2H_6, HCOOH, CH_3OH and C_2H_4 are overestimated compared to the values presented by Johansson et al., 2022. PAN however, agrees over large parts of the flight track well with peak values of 1ppb but is also too high south of -22°S. Johansson et al. 2022 conclude as well that fires were the origin of the observed elevated levels of several organic species. EMAC (and thus CAABA using the same chemical mechanism) generally overestimates VOC mixing ratios over Amazonia ([Pozzer et al., 2022] and discussions therein).

(3) The test with elevated VOC mixing ratios was performed to compensate potentially missed convective events by the Lagrangian trajectory model which transport generally boundary layer air masses into the UT. At the same time it is likely that - by understimating convective uplift - we do not only underestimate the uplift of biogenic species (as assumed in scenario CAPEhigh + noBL) but that we underestimate the uplift of biomass burning species. As EMAC (and CAABA) generally overestimated VOC mixing ratios it is however, difficult to determine the potential strength of missing uplift.

(4) In addition we have to mention (also in the revised version):
in the time range between 18:30 and 19:15 (-22° S to -19° S), air mass history is different compared to the time before and after: Boundary layer contacts in this time took place over south east South America (south of Buenos Aires, see cluster of blue dots in Fig. 3a,b)). Biogenic emissions from there are not as strong as from Amazonia explaining the small difference between scenarios FLASH_{BL} and $\text{FLASH}_{\text{noBL}}$. In this time range, O_3 production for both scenarios is predominantly driven by lightning NO_x. This difference in biogenic emissions fits to the VOC emission distribution (e.g. [Sindelarova et al., 2014] Fig. 2a). However, there were fires over northern Argentina in regions where trajectories had BL contact.

(5) Regarding CO mixing ratios in the upper troposphere, they show a distinct annual cycle with largest values at the end of austral spring (i.e. October). If biogenic emissions would all year round dominate CO mixing ratios, we should not see the observed annual cycle; CO mixing ratios are higher in September and October than in January-March, i.e. the time of year with highest biogenic emissions. In October, the entire upper troposphere is saturated with high amounts of CO. If we use this as a marker for biomass burning subsequently other species should have high fire emissions. At the same time, biogenic emissions are lowest in the South American dry season (July - October). The observations analysed in the [Tripathi et al., 2025] paper were made from December to January, thus at the beginning of the South American wet season where biogenic emissions are significantly larger than during the time of our measurement flights.

(6) The ratio between CO_2 and CO mixing ratios is distinctly different for flights ST06 and ST19: for the first flight both species are negatively correlated with a slope of the regression line of m=-7.8, r^2=-0.65), whereas ST19 is characterised by a positive slope and is higher correlated

(m=8.65, r^2=0.9), as typical for enhanced biomass burning influence. Other studies report slopes of m=0.057 [Mauzerall et al., 1998] and m=40 [Hooghiem et al., 2020] for biomass burning plumes. The ranges of slopes of the regression line between CO_2 and CO is obviously very large. However, in case of a biomass burning impact, the slope is always positive (like for ST19).

(7) We want to reapeat here one of the answers given to a question/remark of reviewer 1:
[Dickerson, 1984] present NO observations taken along the Brazilian coast in December 1983 on board of a commercial aircraft along a similar flight track as for flights ST06 and ST19. They observed smaller mixing ratios of NO in the northern hemisphere (≈ 0-30 ppt) compared to the southern hemisphere (≈ 50-150 ppt, in the latitude range between -25° and 0°S) and attribute this difference to enhanced lightning activity over the South American continent and assume that biomass burning does not play a role for upper tropospheric NO level in December. If this is true, the difference in their observations and our observed NO mixing ratios, being at the same locations more than a factor two higher than reported by Dickerson et al. (1984) provides a further indication that our observations are strongly impacted by biomass burning.

(8) The EMAC model atmosphere is also saturated with fire emitted species in October determining the background. Thus, all trajectories are exposed to these background air masses, also those of scenarios REST and noBL. Therefore, we performed another set of simulations based on an EMAC simulation in which biomass burning emissions were switched off. This leads to O_3 mixing ratios which are ≈ 10 ppb smaller than in our base simulation (Fig. 4b).

Further minor comments:

Line 12: The 50 mWm-2 does not appear anywhere else in this manuscript. Where does this number come from. Please specify in Section 3.5 and/or 4.
Corresponding statements have been added both in Sect. 3.5 and the conclusions and the corresponding number in the abstract has been adjusted.

Line 31: Abbreviation BB emissions is not introduced yet.
We added: biomass burning (BB) emissions.......
Line 198: Abbreviation BL is not introduced yet.
It was introduced in line 177.
Line 89: "NO, NOy" comma missing.
We added the comma.
Line 112: Please add the uncertainty for the NOy and NO measurements and remove double brackets around the citation.
We changed the text in line 112 following your advise.

Line 91: Please specify which parts of these observations were under day- and which under night-time conditions.
We added a second x-axis to the plots in Fig. 2. The observations shown for ST19 were solely made during daytime. For ST06 the shown time range includes a short time before sunrise. The shown time range for flight ST06 is 06:46 UTC to 10:00 UTC and from 17:21 UTC to 20:08 UTC for ST19.

Line 118: "...,it can retrieve vertical profiles for almost two independent layers of CO." This statement on it's one is not very helpful and unprecise. As it is not necessarily needed for the utilization of the MOPITT data in this study, I would suggest removing this part.
We follow your suggestion shortening this section.

Line 182: Is the flash count of the GLM observations used for the estimation of the lighting NOx individually for each timestep or is 0.02 ppb NO per timestep added

for all lightning cases? Please specify.
We give the same answer here as to a similar question by reviewer 1:
We rephrased and shortened the section, explaining now solely how LNO_x emission are treated in our model setup (l.178-183):
If a specific trajectory crosses a lightning event detected by the GLM sensor on board GOES-16 (see Fig. 1), a constant emission of 0.02 ppb NO is added to the NO concentration at this particular CAABA simulation time of the trajectory.

Line 253: As the N2O measurements are not shown anywhere, please quantify the observed range and compare it with literature values for the not specified "tropospheric range".
The comparison with NOAA CMDL flask for Ushuaia in September 2019 show a value of 331.4 ppb_v as the tropospheric value. Consequently, all values below 330.5 ppb_v can be assumed to have stratospheric influence following the same approach of a chemical tropopause definition as defined similarly by e.g., [Müller et al., 2015]. However, as the N_2O values are not shown in the manuscript and they are not relevant for our analysis, we will remove that statement from the revised manuscript version.

Figure 6 caption: 300 hPa values are shown in blue not in yellow.
We changed 'yellow' to 'blue' in the caption.

References

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