Response to Reviewer #3

The study evaluates several available NOx emission inventories from biomass burning and anthropogenic activities using GEOS-Chem sensitivity simulations against a few ground-based measurements and multiple satellite observations in Southern Africa. While the manuscript is readable, it lacks depth in scientific analysis. The conclusions are based solely on sensitivity simulations with altered emissions, ignoring other factors that might affect surface ozone, NOx concentrations, and vertical column densities. The authors seem to imply that GEOS-Chem is flawless except for the input emission inventories, which is obviously untrue. Additionally, when evaluating anthropogenic NOx emissions, the uncertainties of QFED2 are not mentioned, which could undermine the entire analysis. Thus, the current analysis is unconvincing, even if some conclusions might be correct. A more comprehensive analysis is needed to draw more robust conclusions.

Reply: We thank the reviewer for the constructive comments and suggestions, which are very helpful for improving the clarity and reliability of the manuscript.

Firstly, we fully agree with the reviewer that it is hard to quantify the BB and anthropogenic emissions by only evaluating model simulations against satellite observations. Our aim is to understand the key drivers of tropospheric ozone over the understudied Southern Africa by combining model simulation and multiple measurements. To highlight this aim, we have changed the title of this manuscript to "Revisiting the high tropospheric ozone over Southern Africa: role of biomass burning and anthropogenic emissions".

Although there are still some existing limitations in the current version, we have greatly improved the manuscript by (1) comparing the role of BB NO_x and VOC in ozone formation (Figure S6) and confirming the dominated contribution of BB NO_x emissions to simulated ozone difference; 2) highlighting the spatial representativeness of different BB inventories supported by satellite measurements (Figure S4 and Table S1); 3) clarifying the seasonal and vertical dependence of simulated ozone to BB emissions (Figure S3 and Figure S7).

In short, relative to previous work by global models, our study provides a deeper understanding of tropospheric ozone formation from regional to urban scale over the Southern Africa by combining a set of nested chemical transport model simulations and the available measurements. Please find our detailed response (in blue) in the following.

Line 23: Please provide the full words of GFED4.1 at its first appearance, similar to other acronyms throughout the manuscript.

Reply: Added.

Line 34: “high-quality” to “high-resolution”

Reply: Changed.

Line 42-43: What do you mean by the photochemical oxidation of nitrogen oxides? Which species is NO2 oxidized to? Please change “oxidation” to “reactions.”

Reply: We meant the photochemical reactions of NO_x, and we have changed it to “photochemical reactions” in **Line 43**.

Line 55: Delete “emissions” and change “emit” to “emits.”

Reply: Changed.

Line 107: Delete the last “The.”

Reply: Corrected.

Line 130: “there” to “these”?

Reply: Corrected.

Lines 141-144: It would be better to clarify the uncertainties of these satellite datasets.

Reply: Thank you for your suggestion. We have added some discussions on the uncertainties of satellite data in **Lines 153-156**: “Although these satellite datasets have been well employed to reflect emission changes, their uncertainties are also notable due to biases in slant column density, air mass factor, and stratosphere-troposphere separation. For example, the reported uncertainties in NO_2 columns from OMI and TROPOMI are 25-50% and they can be increased to 50-100% in terms of OMI HCHO columns.”.

Lines 167-189: Why are these sensitivity simulations conducted in different years? Are you sure these

simulations are consistent, considering the natural variability of climate? In Line 169, you mentioned simulations from 2019-2023, but I didn't find any corresponding simulations in Table 1.

Reply: Considering the availability of observation data, we conducted these sensitivity simulations for different years with varying emissions and meteorology. But we didn't run the model consecutively from 2019 to 2013. To avoid misunderstanding, we have revised the manuscript with the following updates:

Line 184: "Here we focused our experiments on July-August 2019 for all sensitivity and benchmark simulations."

Lines 186-188: "Firstly, we used GFED4.1 and QFED2 inventories to simulate the hourly air pollutant concentrations for July-August, 2019 (Run_GFED and Run_QFED), respectively, and validated the model results with satellite observations."

Line 195: "Considering that surface air pollutant measurements are only available for June-August 2023, and then …"

Figure 2. Why didn't you use soil NOx and BVOC emissions from your simulations but those from Offline documents? Soil NOx and BVOC are sensitive to meteorological conditions and are calculated online in GEOS-Chem.

Reply: In the GEOS-Chem model, the offline soil NO_x and BVOC emission inventories that were archived from previous model runs are the same with our online calculated emissions.

We have revised the caption of Figure 2: "Anthropogenic NO_x and VOC are from CEDSv2 inventory, soil NO_x and BVOC are calculated by the GEOS-Chem model, and biomass burning NO_x and VOC are from GFED4.1 and QFED2 inventory."

Lines 234-235: Compared to what? The observed 70 ppb? Do the simulation results and observations match in timing?

Reply: The observation time for the Rwanda site is 2015-2017, and 70 ppb is the daily maximum ozone concentration at that observation time. Our simulation period is July-August 2019. We have noted this in **Line 255**: "Compared to the observed ozone in Rwanda, it may indicate…".

Lines 238-239: I don't understand the logic. How can the simple comparisons above support such a conclusion? How about the surface ozone seasonal variations? Are surface ozone concentrations lower

in the non-fire season?

Reply: Surface ozone in this region is characterized by strong seasonal variation with high values in the fire season and low values in the non-fire season, supported by model simulations and ozonesonde measurements: 1)We compared the simulated surface ozone concentrations during the fire season (July-August 2019) with those during the non-fire season (January-February 2020) in **Figure S3**. It shows the high ozone of 70 ppb during the fire season is over Southern Africa, while during the non-fire season ozone concentrations in Southern Africa are only around 35 ppb. 2) Based on the vertical distribution of seasonal ozone over Ascension Island, UK in 2018, the seasonal enhancement is evident with the highest summer ozone in the lower atmosphere (**Figure R1**).

To clarify this argument, we have added in **Lines 267-269**: "Considering the strong seasonal variation of surface ozone in Southern Africa (**Figure S3**) and the estimated ozone precursors from different sources in Figures 3, here the large differences in simulated surface ozone with different BB inventories demonstrate that BB contributes greatly to high ozone concentrations during the fire season in Southern Africa."

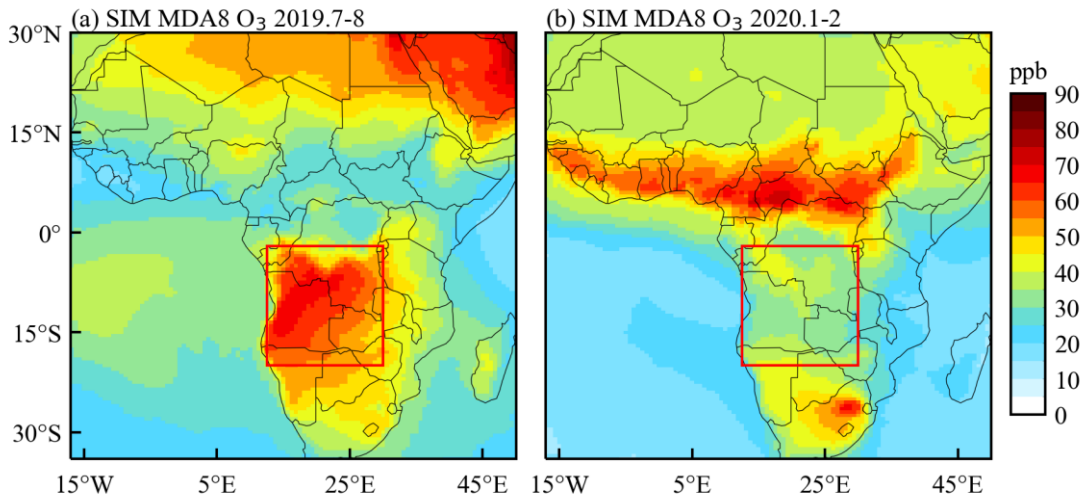

Figure S3. Spatial distribution of surface ozone during the fire season (July-August 2019) and non-fire season (January-February 2020) simulated by GEOS-Chem.

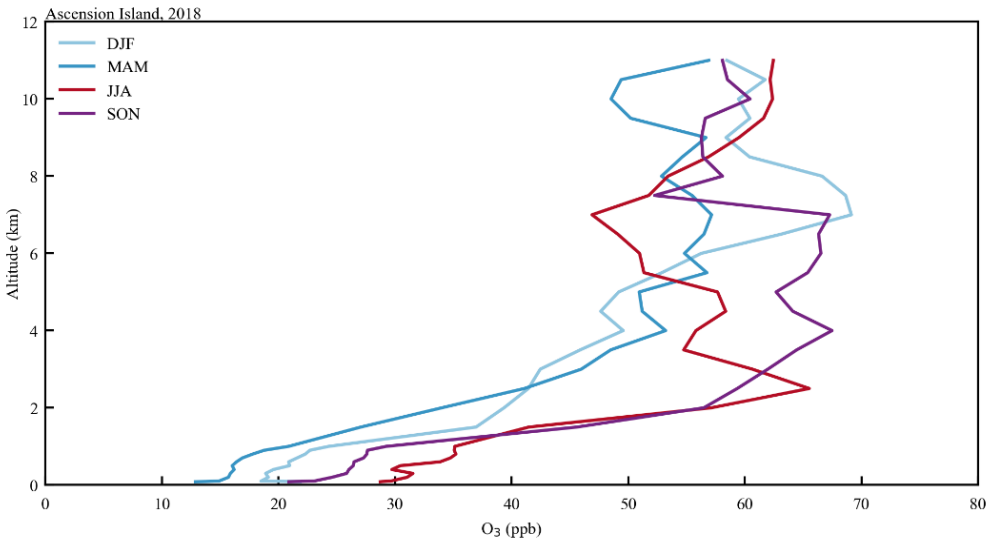

Figure R1. Vertical distribution of seasonal ozone over Ascension Island, UK, 2018.

Lines 244-245: I wonder how you processed the observed and modeling data. Aren't coincident observations and model results used in the comparison? Did you just calculate the seasonal or monthly mean model values regardless of the availability of observations?

Reply: We sampled the model data with the same satellite overpass times for comparison and excluded the simulation results with missing satellite measurement.

We noted this point in **Lines 152-153**: "We sampled the model simulation results consistent with satellite overpass times in the following comparisons."

Line 263: It can only explain the lower NOx emissions of FINNv1.5. How about GFAS?

Reply: We have added this discussion for GFAS in **Lines 288-290:** "This lower estimate in the bottom-up FINNv1.5 inventory may be attributed to the underestimated burned area and emissions (Wiedinmyer et al., 2011), and the lower top-down GFAS estimate could be due to a smaller emission factors (Liu et al., 2020)."

Line 271: "he" to "the".

Reply: Corrected.

Line 271-272: You can't make such a conclusion based on a single set of sensitivity tests with perturbed BB NOx emissions, although the conclusion may be correct.

Reply: We agree with the referee and have revised this argument to in **Lines 311-314**: "This remarkable discrepancy suggests that the uncertainties in BB emissions could play an important role in simulating surface ozone over Southern Africa. This is consistent with previous work that BB emissions lead to strong ozone increases in Southern Africa during the fire season (V. Clarmann et al., 2007; Jaffe and Wigder, 2012)."

Line 276: Delete "whether" and add "simulating" before "in."

Reply: Changed.

Line 282-283: The differences are also significant in the mid-troposphere. Did you calculate which part in altitude contributes most to the TCO difference between GFED4.1 and QFED2? It can't be directly derived from the vertical profiles in Figure6.

Reply: Thanks for the comment. Yes, as seen in Figure S6, the difference in tropospheric ozone concentrations between GFED4.1 and QFED2 is also notable at 3-6 km. This altitude is also the maximum contributor to TCO in this region. The ozone vertical profiles in Ascension Island simulated by the two BB inventories in Figure 6 also differ notably at this altitude range.

We have revised the text in **Lines 326-328**: "The differences in ozone vertical distribution due to the two BB inventories are notable in the troposphere below 6 km, in particular at the altitude range of 3-6 km (Figure S7). Compared to the ozonesonde observations, this bias can be also found while GEOS-Chem captures the vertical ozone variations well regardless of which inventory is used. This is consistent with the results of small TCO differences in Figures 5a-5b."

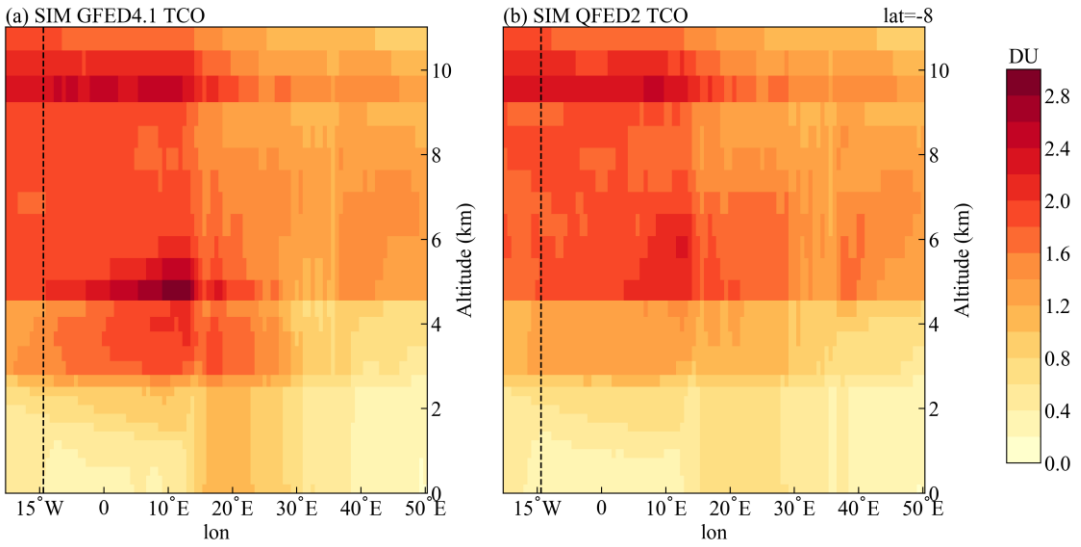

Figure S7. Vertical profiles of column concentrations across the latitude of -8°S simulated by GEOS-Chem using the GFED4.1 and QFED2 inventories, with the dashed line indicating the location of Ascension Island.

Line 296-305: NO2 vertical column retrieval is sensitive to the NO2 vertical profile. Did you redo the retrieval using your simulated NO2 vertical profiles?

Reply: Thanks for your suggestion. Due to the multiple satellite products, we didn't particularly recalculate the NO_2 columns using the GEOS-Chem simulated vertical profiles. We have pointed out this issue in **Lines 350-351**: "It is noted that this comparison between the simulated and satellite-based tropospheric columns could be biased due to their different representativeness in vertical profiles of chemical species."

Line 410-411: I'm afraid I must disagree with such a derivation.

Reply: We have removed this argument.

Line 454: "high" to "large low"

Reply: Changed.

Line 467-468: I don't understand the logic here. The sea downwind is also susceptible to BB's long-range transport.

Reply: Contribution from anthropogenic sources to NO_2 columns in Luanda should be reduced during

the fire season, so we attempted to attribute the smaller underestimation of NO_2 in Luanda during fire season to the long-term transport of BB emissions. To make it clearer, we have reorganized the text as follows:

Lines 535-537: "Due to the decreased contribution from anthropogenic sources to NO_2 columns during the fire season, the moderate underestimation during fire season (July-August 2019) in Luanda may be due to the long-term transport of pollutants from biomass burning to urban areas."

Line 472: "like Southern Africa" to "with significant NOx spatial heterogeneity."

Reply: Changed!

Lines 493-494: Did you check NO2 in the free troposphere? A large portion of NO2 lies in the free troposphere, significantly contributing to NO2 VCD.

Reply: Following your suggestion, we plotted the vertical NO_2 profiles simulated by GEOS-Chem for during fire season and non-fire season (**Figure S15**). During the non-fire season, NO_2 concentrations are only below 1.5 km; during the fire season, the important contribution to NO_2 columns can extend around 7 km. As such, for the analysis in this section of Figure 12, we selected March-April 2018-2023 for comparison in order to exclude the effect of biomass burning and to more accurately reflect the bias of anthropogenic emission inventories in the region.

We have added this point in **Lines 361-363**: "The tropospheric NO_2 during non-fire season is dominantly contributed by the lower atmosphere (Figure S14), and then we selected March-April 2018-2023 for comparison in order to exclude the effect of biomass burning."

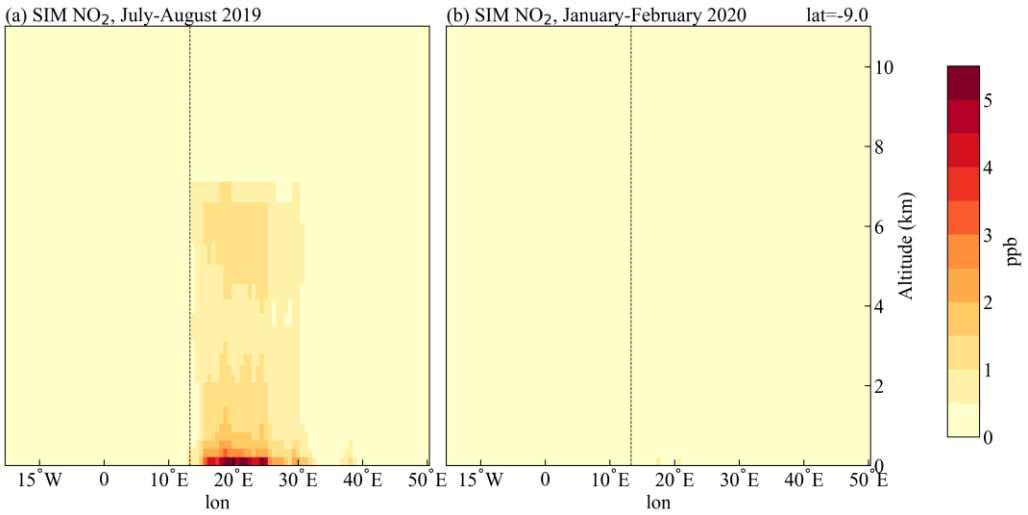

Figure S14. Vertical profiles of NO_2 across the latitude of -9°S (Luanda is located at 8°80'S, 13°23'E) simulated by GEOS-Chem during the fire season (July-August 2019) and non-fire season (January-February 2020), with the dashed line indicating the location of Luanda.

References:

Jaffe, D. A. and Wigder, N. L.: Ozone production from wildfires: A critical review, Atmospheric Environment, 51, 1-10, 10.1016/j.atmosenv.2011.11.063, 2012.

Liu, T., L.J. Mickley, M.E. Marlier, R.S. DeFries, M.F. Khan, M.T. Latif, and A. Karambelas (2020). Diagnosing spatial biases and uncertainties in global fire emissions inventories: Indonesia as regional case study. Remote Sens. Environ., 237, 111557.

v. Clarmann, T., Glatthor, N., Koukouli, M. E., Stiller, G. P., Funke, B., Grabowski, U., Hopfner, M., Kellmann, S., Linden, A., Milz, M., Steck, T., and Fischer, H.: MIPAS measurements of upper tropospheric C_2H_6 and O_3 during the southern hemispheric biomass burning season in 2003, Atmospheric Chemistry and Physics, 7, 5861–5872, https://doi.org/10.5194/acp-7-5861-2007, 2007.