Response to Reviewer #2

In this study, the authors integrated the high-resolution GEOS-Chem model and newly-available measurements to estimate the impact of biomass burning (BB) and anthropogenic emissions on tropospheric ozone over Southern Africa. They identify the best estimate of BB emissions inventory and quantify the effect on regional tropospheric ozone over Southern Africa. The authors compare simulation outputs using different emission inventories. However, the discussion would be strengthened by providing a more in-depth coverage of the physical and chemical processes driving ozone and PM formation.

Reply: Thank you very much for your suggestion. We have carefully addressed the comments and the point-by-point responses are in blue.

Comments:

1. The authors should provide a summary of available surface observations and satellite data for chemical species and compare the model results with observations, including statistics on the spatial distribution.

Reply: Thank you very much for your suggestion.

We have added the surface observation data and the satellite data used in Table 1. Meanwhile, we compared the simulation results against satellite observations in Table S1, and the details of each surface site was described in the text.

Table 1. Satellite and surface observations used in this study.

Species		Spatial resolution/ site locations	Observation period
O_3	OMI	1 ° × 1.25 °	July-August 2019
NO_2	OMI	0.25 ° × 0.25 °	2019-2020
HCHO	OMI	0.05 ° × 0.05 °	July-August 2019
NO_2	TROPOMI	0.125 ° × 0.125 °	2018-2023
AOD	MODIS	1 ° × 1 °	July-August 2019
CO	MOPITT	1 ° × 1 °	July 2019

$PM_{2.5}$	Humpata	(14 °34' S, 13 °26' E)	June-August 2023
	Luanda	(8 °48' S, 13 °14' E)	
	Luena	(11 °45' S, 19 °54' E)	
	Lusaka	(15 °24' S, 28 °17' E)	
NO_2	Humpata	(14 °34' S, 13 °26' E)	June-August 2023
	Luanda	(8 °48' S, 13 °14' E)	
	Luena	(11 °45' S, 19 °54' E)	
O_3	Ascension Island	(7 °58' S, 14 °24' W)	July-August 2017-2019

Table S1. Statistics of spatial correlation coefficients between model simulation results and satellite data.

	GFED4.1		QFED2	
	NMB	R	NMB	R
OMI O_3	-10.4%	0.82	-12.9%	0.87
OMI NO_2	22.0%	0.83	9.3%	0.92
OMI HCHO	-2.9%	0.79	-5%	0.76
TROPOMI NO_2	8%	0.78	-3%	0.91
MODIS AOD	-34%	0.9	5.7%	0.89
MOPITT CO	-17.4%	0.89	-17.1%	0.89

2. In addition to the overall emission rate difference, how do spatial variations compare across the different emission inventories? A summary of statistics analysis would be helpful.

Reply: Thanks for this very helpful suggestion. We have compared their spatial differences in Figure S1 and some description in **Lines 293-300:**

"Spatially, there are also evident differences among different biomass burning inventories (**Figure S4**). The spatial distribution of the high values in GFED4.1 and QFED2 is generally consistent with a spatial correlation coefficient of 0.76, both showing high emissions in northeastern Angola. In contrast, the GFED5 inventory has high NO_x emissions concentrated in southwestern Congo, and its spatial

distribution differs considerably with QFED2. The GFAS inventory has a similar spatial distribution with QFED2 (a correlation coefficient of 0.84), but GFAS cannot capture the localized high emissions as shown in QFED2 and GFED4.1. However, the FINNv1.5 and FINNv2.5 exhibit a very different spatial distribution compared to other inventories, with low emissions in Angola and high emissions in the Congo region. Their spatial correlation coefficients with the QFED2 inventory are 0.06 and 0.31, respectively."

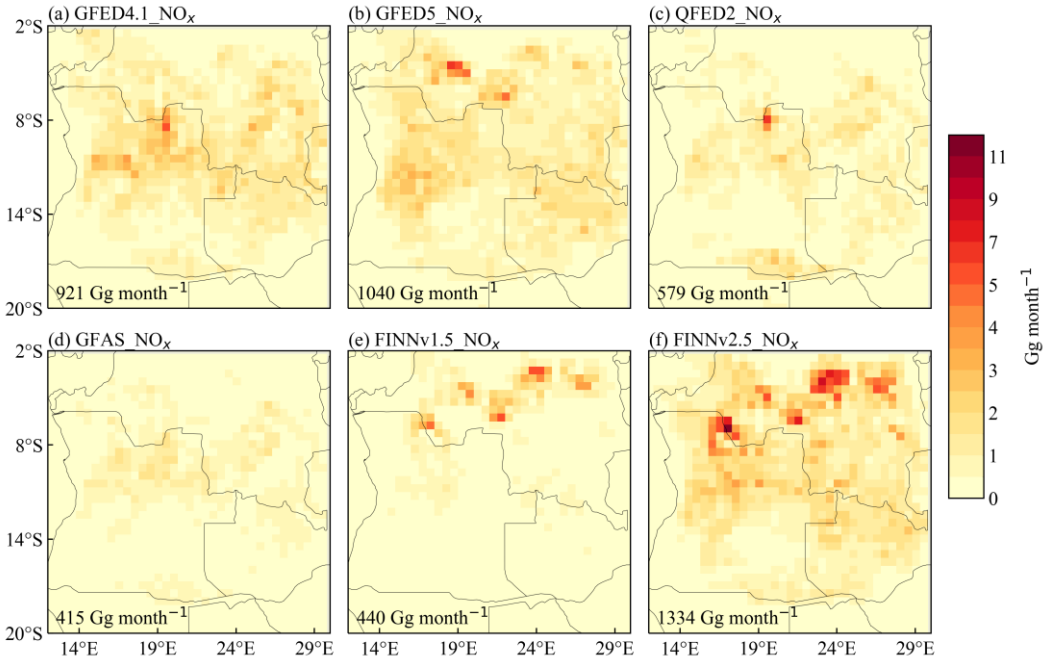

Figure S4. Spatial distribution of monthly NO_x emissions from different biomass burning emission inventories in July-August 2014.

3. BB not only emits NOx, but also VOCs and PM. The authors should summarize the related information such as CO, VOC, NOx, BC and OC, which were stated to play a role in ozone concentration. Are there any specific ratios among emitted chemical species?

Reply: We calculated the total CO, VOC, NO_x, BC and, OC emissions from the six BB inventories in **Figure S5** and have added them in **Lines 302-305**: "In addition to NO_x emissions, the VOC emissions are the highest in GFED5 and FINNv2.5 inventories, and the other four inventories show much smaller VOC emissions. Each inventory adopts different specific ratios for emitted chemical species, but they also differ with each other. For example, there is a NO_x/OC ratio of 1:0.6 in GFED4.1, 1:1.5 in GFED5, GFAS, and FINNv1.5, 1:3 in QFED2, and 1:1 in FINNv2.5 (Figure S5)."

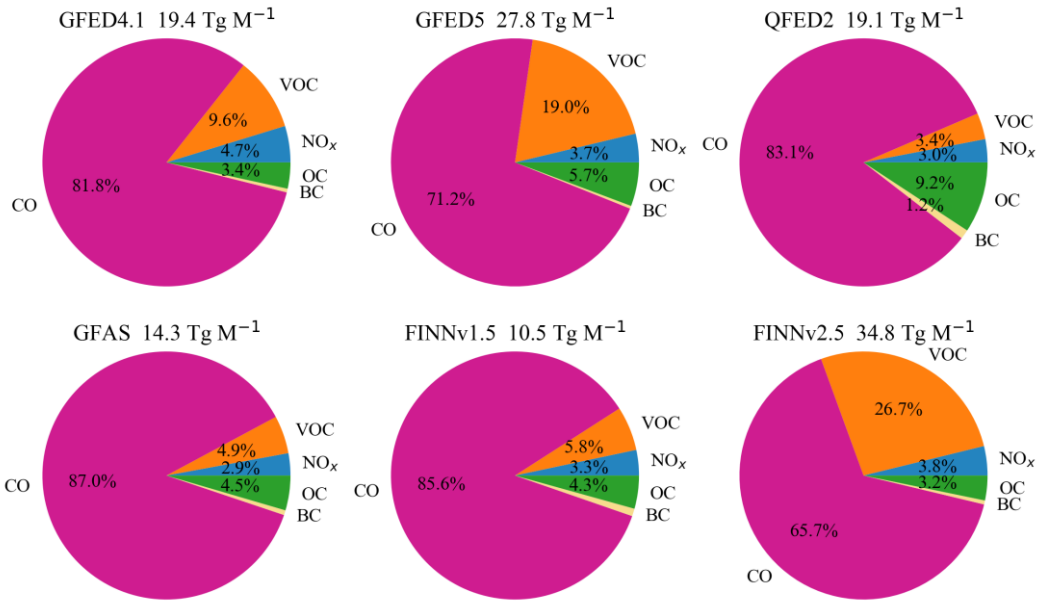

Figure S5. Estimated species emissions (Tg month^{-1}) in different biomass burning emission inventories in Southern Africa, July-August 2014.

4. Line 174: should "Run_QFED_34%" be corrected to "Run_QFED_66%NOx"?

Reply: Corrected.

5. Lines 227-228: When comparing ozone concentrations with Dewitt et al. (2019), the authors should present results for both GFED and QFED emissions at the grid point associated with the station location. Currently, only Run_GFED is presented.

Reply: We have added this result in **Lines 254-255:** "Based on our simulation results, it can be found that the daily maximum ozone during the BB season is 86 ppb for Rwanda in the Run_GFED run, compared to only 62 ppb in Run_QFED run."

6. Lines 235-240 and Figures 5(a)-(c): OMI O3 shows significantly higher ozone concentrations over the Atlantic Ocean compared to the simulation. Could this discrepancy be related to the meteorological conditions in the model? This issue might also influence the comparison of NOx concentrations between the model and observations in the studied cases.

Reply: This discrepancy is not caused by the MERRA-2 reanalysis meteorology in the model. And we

show that the higher OMI ozone concentrations over the Atlantic Ocean is mainly due to the different background ozone levels between the satellite and model simulation.

We have discussed this in **Lines 259-260**: "Also, in Figure S2, we find that the GEOS-Chem simulated (Run_QFED) and OMI tropospheric ozone columns are in good agreement over the Atlantic Ocean after individually subtracting the background ozone values."

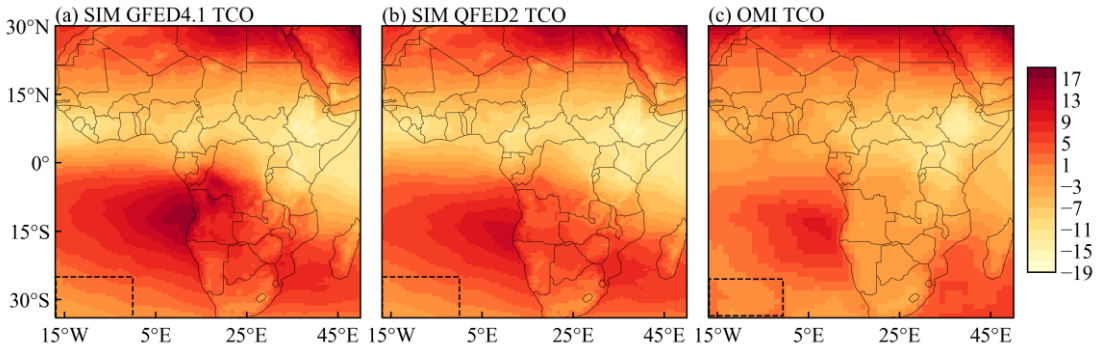

Figure S2. The comparison of GEOS-Chem simulated (left and middle panels) and satellite-based (right panel) tropospheric ozone columns after individually subtracting the background ozone values averaged over the black box (-34~-25°S, 17°W~0).

7. Line 303: if the case with QFED2 NOx emissions reduced by 34% (Figure S3) better aligns with satellite TCO data, would the FINNv1.5 emission inventory, which has ~ 0.67 of QFFD2 NOx emission (Figure 4c), be a more appropriate NOx inventory for this study?

Reply: No, the FINNv1.5 inventory wouldn't be more appropriate due to its strong spatial biases. Although the FINNv1.5 NO_x emissions are similar to the 67% of QFED2 NO_x emissions, in **Figure S3** it can be found that the spatial distribution of the FINNv1.5 NO_x emissions is quite different from other inventories, e.g., with a spatial correlation of 0.06 with the QFED2 inventory and of 0.29 with GFED4.1 inventory. After evaluating with the satellite NO_2, we demonstrate that the QFED2 inventory has a better regional representativeness of NO_x emissions, as shown in **Table S1**.

8. Figures (g)-(l): the authors should address why the model predicts relatively low HCHO and CO concentrations.

Reply: We have explained this in the revision as follows:

Lines 362-364: "and the underestimated HCHO columns in GEOS-Chem might be due to some missing VOC species (Zhao et al., 2024) and the lower anthropogenic NO_x emissions in Southern Africa that both affect the chemical production of HCHO."

Lines 375-377: "The regional average of CO column concentrations simulated by GEOS-Chem is underestimated by approximately 10% compared to MOPITT, which reflects a long-lasting issue of CO underestimation in GEOS-Chem model (David et al., 2019; Ni et al., 2018)."

9. Since pollutant concentrations can exhibit strong diurnal variation, was the simulation data aligned with the satellite overpass times in the region for the model-observation comparison?

Reply: Yes! We did sample the simulation results with the satellite overpass times, and have added this point in **Lines 152-153**: "We sampled the model simulation results consistent with satellite overpass times in the following comparisons."

10. Lines 318-319: how do BB VOC emissions in both emission inventories compare with anthropogenic (AVOC) and biogenic (BVOC) VOCs in Figure 3?

Reply: We have included BB VOC emissions in Figure 3, and added some comparison in **Lines 234-236:** "BB VOC has similar seasonal variability in both inventories, but the GFED4.1 inventory emits 2-3 times as much as the QFED2 inventory in fire season. The BVOC emissions are generally higher than BB VOC emissions except for those in July-August months from the GFED4.1 inventory."

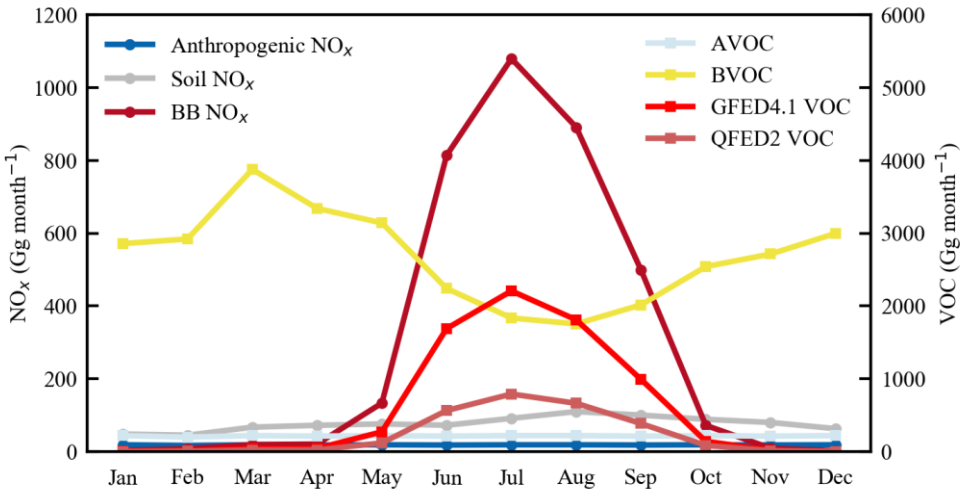

Figure 3. Seasonal variations in anthropogenic NO_x (deep blue), soil NO_x (grey), biomass burning

NO_x (red), biomass burning VOC (red), anthropogenic VOC (blue), and biogenic VOC (yellow) emissions in 2019 (unit: Gg $month^{-1}$). 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.

11. Lines 327: What are the major chemical species in BB VOCs, and how do they influence ozone formation beyond HCHO formation?

Reply: According to **Figure R1**, the major chemical species from BB VOC are OVOC, followed by alkanes, and alkenes; and OVOC and alkenes dominate the chemical formation of ozone and HCHO. However, we find the impact of uncertainties in BB VOC emission in ozone formation is much smaller than that from BB NO_x emissions (Figure S6).

We have added this in **Lines 352-355**: "In contrast, we find that the BB VOC emissions from GFED4.1 inventory are about 3 times the QFED2 inventory in fire season, but the regional mean changes are only 2.5 ppb for MDA8 ozone and 0.94 DU for TCO for July-August 2019 in response to a tripled QFED2 VOC emissions (Figure S6)".

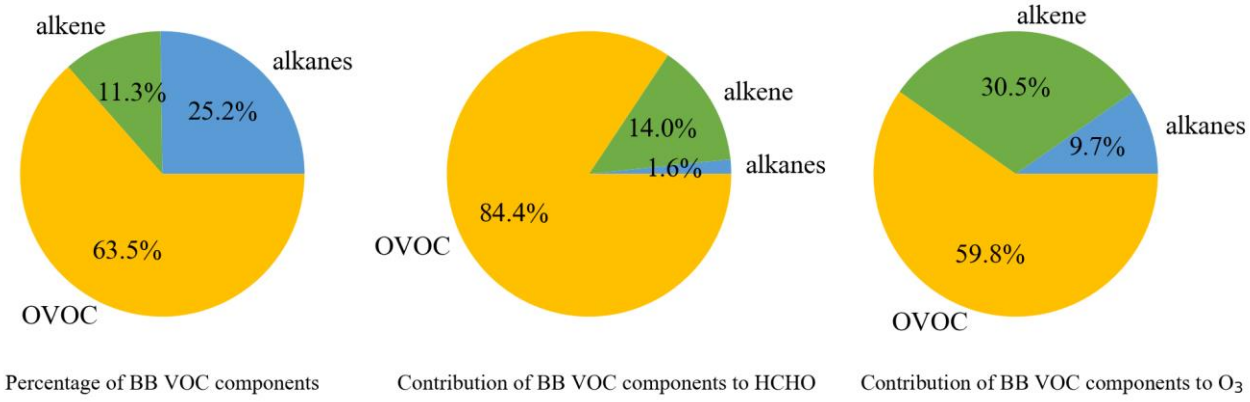

Figure R1. NMVOC components of the QFED2 inventory and their percentage contribution to formaldehyde and ozone.

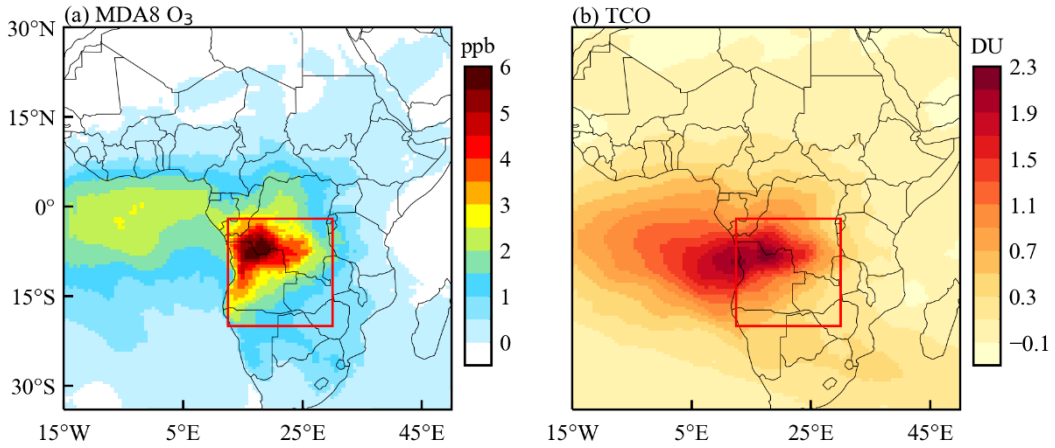

Figure S6. Changes in surface MDA8 ozone (left) and TCO (right) when VOC emissions from QFED2 inventories are tripled for July-August 2019. The regional mean changes are 2.5 ppb for MDA8 ozone and 0.94 DU for TCO.

12. Lines 340-341: The authors briefly mention the model results without adequate discussion. A more detailed explanation of how aerosol chemical processes influence surface ozone concentrations would be helpful to illustrate the causality.

Reply: We have added the following discussion in **Lines 394-397:** "Aerosol chemistry mainly influences ozone formation by altering photolysis and heterogeneous processes. On the one hand, aerosol can change the shortwave radiation reaching the ground through scattering and absorption, which in turn affects the photolysis rate. On the other hand, aerosol can update reactive radicals (e.g., HO_2, nitrogen radicals) that are critical for ozone formation."

13. Figure 9 and the associated discussion: The authors should evaluate the comparison between observations and simulations. Could the higher observed NOx concentrations at the observation site compared to the simulation be due to the emissions being concentrated in a small area, whereas the model averages emissions over a larger grid? This could explain the lower simulated concentrations.

Reply: Yes. The location of the four surface observation sites is shown in Figure R2. The stations are primarily located alongside streets. As you mentioned, the observed pollutant emissions are concentrated in smaller areas, whereas the model averages the emissions over a larger grid, which is one of the reasons for the underestimation of the modeled results compared to the observations,

We have added this point in **Lines 458-459**: "although the lack of model resolution accuracy is also a reason for the underestimation at the station scale."

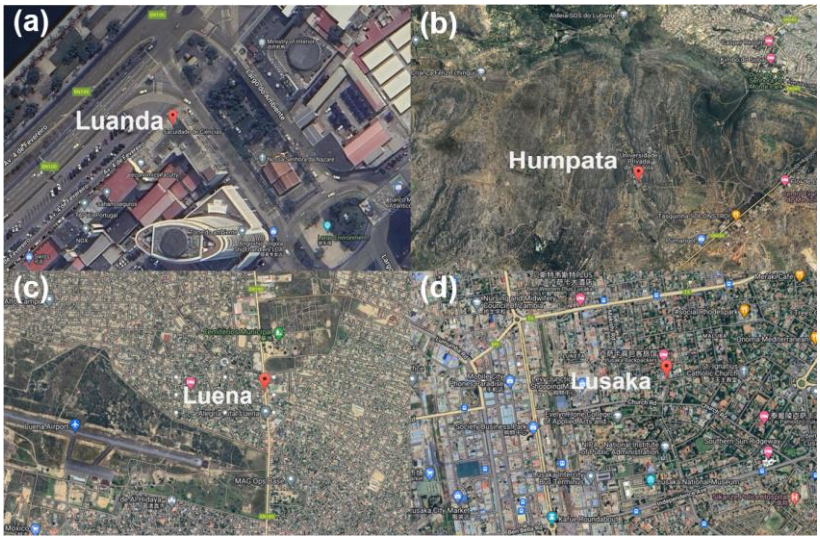

Figure R2. Location of four surface observation sites (Google Earth).

References:

David, L. M., Ravishankara, A. R., Brewer, J. F., Sauvage, B., Thouret, V., Venkataramani, S., and Sinha, V.: Tropospheric ozone over the Indian subcontinent from 2000 to 2015: Data set and simulation using GEOS-Chem chemical transport model, Atmospheric Environment, 219, 10.1016/j.atmosenv.2019.117039, 2019.

Ni, R., Lin, J., Yan, Y., and Lin, W.: Foreign and domestic contributions to springtime ozone over China, Atmospheric Chemistry and Physics, 18, 11447-11469, 10.5194/acp-18-11447-2018, 2018.

Zhao, T., Mao, J., Ayazpour, Z., González Abad, G., Nowlan, C. R., and Zheng, Y.: Interannual variability of summertime formaldehyde (HCHO) vertical column density and its main drivers at northern high latitudes, Atmospheric Chemistry and Physics, 24, 6105-6121, 10.5194/acp-24-6105-2024, 2024.