



## Supplement of

## **Revisiting the high tropospheric ozone over southern Africa:** role of biomass burning and anthropogenic emissions

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**Figure S1.** GEOS-Chem modelled surface ozone concentration obtained from the July 2019 benchmark experiment (downloaded from https://ftp.as.harvard.edu/gcgrid/geoschem/1mo\_benchmarks/).



**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).



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



**Figure S5.** Estimated species emissions (Tg month<sup>-1</sup>) in different biomass burning emission inventories in Southern Africa, July-August 2014.



**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.



**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.



**Figure S8.** Comparison of the GEOS-Chem simulated NO<sub>2</sub> columns in Africa in July-August 2019 using the QFED2 inventory (a) with OMI (b) and TROPOMI (c). Numbers below the red boxes indicate the regional averages.



**Figure S9.** GEOS-Chem simulated surface ozone, tropospheric ozone columns, and NO<sub>2</sub> columns in Africa for July-August 2019 and its comparison with satellite data. The left panels are results from the baseline simulation; the middle panels are results from the simulation with 34% reduction in NO<sub>x</sub> emissions from the QFED2 inventory; the right panels are results from the satellite data.



**Figure S10.** The simulated effects of aerosol chemistry on MDA8 ozone (top) and tropospheric ozone columns (bottom) in July-August 2019 by using the GFED4.1 and QFED2, respectively.



**Figure S11.** Time series of the simulated and observed (black) median daily  $PM_{2.5}$  concentrations for June-August 2023. a-d are for Humpata, Luanda, Luena, and Lusaka, respectively. The plots labelled by the "SIM\_noDST" and "SIM\_10NO<sub>x</sub>" are the  $PM_{2.5}$  concentration after removing dust aerosols and the  $PM_{2.5}$  concentrations when anthropogenic NO<sub>x</sub> emissions were increased by a factor of 10 in CEDSv2.



**Figure S12.** Time series of simulated June-August 2023 surface MDA8 ozone concentrations from the baseline simulation (black) and the sensitivity simulation (red) in which anthropogenic  $NO_x$  emissions were increased by a factor of 10 in CEDSv2.



**Figure S13.** The observed and simulated tropospheric NO<sub>2</sub> columns in June-August 2023. (a) the baseline simulation with QFED2 inventory. (b) the sensitivity simulation with anthropogenic NO<sub>x</sub> emissions increased by a factor of 10. (c) TROPOMI data. (d-f) the sensitivity simulations with sectoral NO<sub>x</sub> emissions increased by a factor of 10 in energy, industry, and transportation. The numbers in the plots are all the relative NO<sub>2</sub> columns enhancement in Luanda area. The dashed boxes indicate the downwind background area, which is subtracted to obtain the relative NO<sub>2</sub> column concentration in Luanda.



**Figure S14.** Vertical profiles of NO<sub>2</sub> across the latitude of -9 °S (Luanda is located at 8 °O'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.



**Figure S15.** The simulated contributions to surface ozone in July-August 2019 from different sources, including biomass burning emissions (left), natural emissions (middle), and anthropogenic emissions (right), under Run\_QFED simulation (top) and under Run\_QFED\_Anth10NO<sub>x</sub> simulation (bottom) where anthropogenic NO<sub>x</sub> sources were increased by a factor of 10. Here the natural emissions refer to the biogenic VOC and soil NO<sub>x</sub> emissions.



Figure S16. Trends in NO<sub>x</sub> emission rates from anthropogenic sources under different future scenarios (Unit: kg m<sup>-2</sup> s<sup>-1</sup>).

|                         | GFED4.1 |      | QFED2  |      |
|-------------------------|---------|------|--------|------|
|                         | NMB     | R    | NMB    | R    |
| OMI O <sub>3</sub>      | -10.4%  | 0.82 | -12.9% | 0.87 |
| OMI NO <sub>2</sub>     | 22.0%   | 0.83 | 9.3%   | 0.92 |
| OMI HCHO                | -2.9%   | 0.79 | -5%    | 0.76 |
| TROPOMI NO <sub>2</sub> | 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 |

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