Measurement of the tropospheric vertical columns of NO_2 and H_2CO over Kinshasa: comparison with TROPOMI and the GEOS-Chem model

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MOTIVATION

Many African cities are affected by the problem of air pollution as mentioned in the study by Louisse et al 2014 stating that air quality in African urban areas is expected to deteriorate in the coming decades.

The city of Kinshasa, the capital of the Democratic Republic of Congo (DRC), a large megalopolis of 12 million inhabitants, with an estimated population of 30 million by 2030 UN (2016), is not spared by this air pollution as shown in McFarlane et al. (2020) and WHO reports. It is dominated by a multiplication of motorcycles, old vehicles, open garbage cans, unpaved roads and the use of embers from forest wood as energy for cooking. Its surroundings are also affected by seasonal forest burning, which is a potential source of several pollutants, including NO_2 and H_2CO.

However, measurements are lacking, and this limits the number of studies address this topic in this region of the world. Even with regard to the worldwide distribution of atmospheric measurement stations (e.g. via the NDACC network www.ndacc.org), it can be seen that Central Africa is largely under-sampled compared to other land areas of the globe. Although satellite observations exist in this region, they are generally unsuitable for sampling over heavily polluted areas due to their low sensitivity near the surface, and the consequences can be directly related to the inaccuracy of the corresponding emission estimates by top-down approaches. Ground-based measurements are and will remain essential for atmospheric research, whatever the future progress of satellite instruments and the refinement of models. Therefore, measurements in this region are of particular interest in order to know the temporal and spatial distribution of NO_2 and H_2CO emission intensity at the local scale and also in order to understand the accuracy of satellites and models.

It is in this perspective that the present work is inscribed, with the objective of presenting a series of atmospheric measurements of NO_2 and H_2CO, going from November 2019 to July 2021, with as vision: 1. to make a validation of the TROPOMI satellite, 2. to evaluate the performance of the GEOS-Chem model to constrain the emissions in this area.

Site description and Instrumental setup

The MAX-DOAS instrument in Kinshasa is installed on the roof of the Faculty of Sciences building at the University of Kinshasa (UniKin: -4.42° S, 15.31° E), which is about 15 m above ground (altitude of about 315 m MSL). The UniKin site is located about 5 km from downtown Kinshasa and about 10 km from the Congo River. More details on the city of Kinshasa and its characteristics are described in Yombo Phaka et al. (2021). The location of the MAX-DOAS instrument is shown in Figure 1. The instrument consists of a scanning telescope, motorized by a stepper motor controlling the elevation angles (0°, 1°, 2°, 3°, 4°, 5°, 6°, 7°,8°,15°,30°,45°,88°) as shown in Figure 1. The scattered sunlight collected by the telescope is redirected to the Avantes spectrometer located in the main housing of the instrument via a 600 µm optical fiber. This spectrometer covers covers a UV-Vis wavelength range (290-550 nm) and its spectral resolution of full width at half maximum is 0.7 nm (FWHM). An on-board computer (PC104) ensures the operation of the whole system (automatic recording of spectra and control of the telescope). The whole system is powered by 220 V/AC.

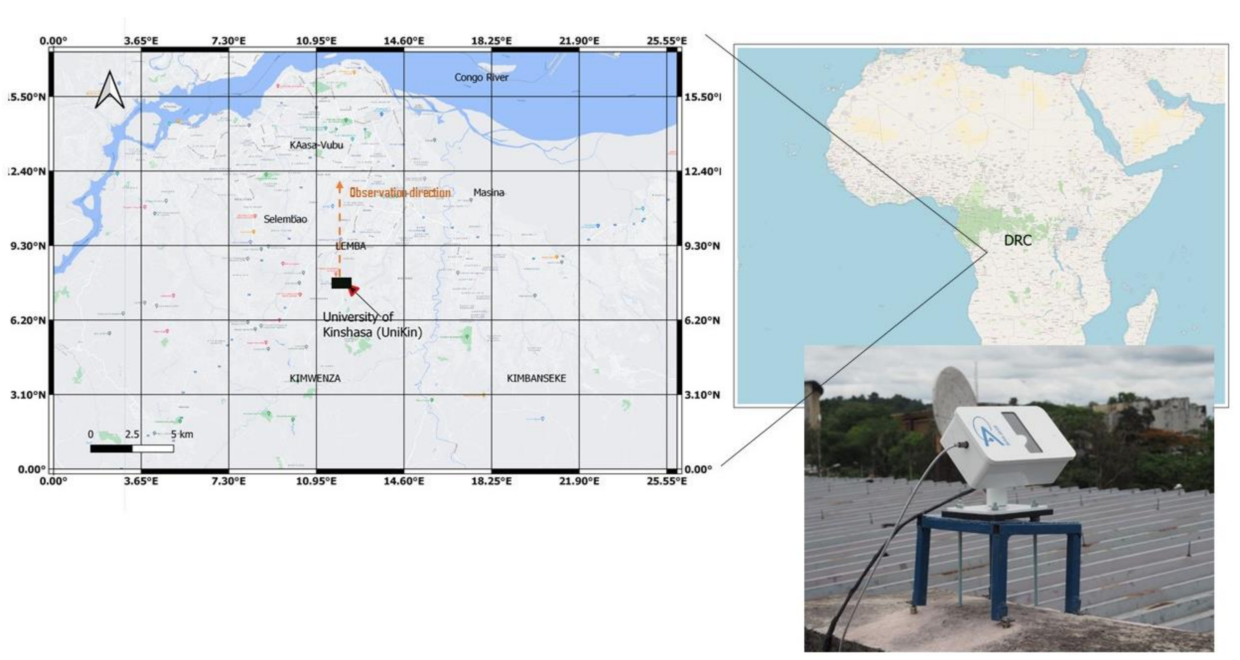

Figure 1. MAX-DOAS instrument installed on the roof of the Faculty of the University of Kinshasa since November 2019

DATA SETS AND METHODS

MAX-DOAS data

The retrieval of NO_2 and H_2CO tropospheric vertical columns densities is performed using inversion tools recently developed in the FRM4-DOAS project (Fiducial Reference Measurements for Ground-Based DOAS Air-Quality Observations) (https://frm4doas.aeronomie.be).

FRM4-DOAS is an international project aiming at harmonizing and standardizing the processing chain of observations from different MAXDOAS stations operated within the International Network for the Detection of Atmospheric Composition Change (NDACC). It consists in an automated processing of spectral data recorded by the instrument (solar radiance and meta data) in near-real time until the production of geophysical quantities used in the GEO/CEOS quality assurance framework for Earth observation (http://qa4eo.org) such as tropospheric and stratospheric NO_2 vertical profiles, total O_3 columns, and tropospheric H_2CO profiles, aerosol extinction and optical depths.

GEOS-Chem model data

This work is based on a standard full chemistry simulation performed with the Goddard Earth Observing System chemistry (GEOS-Chem) model. We use version 12.0.2 (https://doi.org/10.5281/zenodo.1455215) runs implementing MERRA-2 assimilated meteorological fields at a horizontal resolution of 2° × 2.5° (latitude/longitude) on a vertical grid of 72 levels, up to 0.01 hPa (about 80 km). The outputs of the GEOS-Chem simulations are first performed on the vertical grid of each dataset (TROPOMI and MAX-DOAS) and then smoothed using the Averaging Kernel corresponding to each dataset. Our simulation includes EDGAR v4.3 for fossil fuel emissions, EMEP and NEI2011 for regional anthropogenic emissions, GFED v4 for fire emissions, MEGAN v2.1 for biogenic emissions, and RETRO for Non-Methane Volatile Organic Compounds (NMVOCs) emissions. In particular, the Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa) inventory has been used to provide African anthropogenic emissions (Marais and Wiedinmyer, 2016).

TROPOMI data

We exploit reprocessed (RPRO: v0102 for NO_2 ; v1.1 for H_2CO) and off-line (OFFL: v0102 and v0103 for NO_2; v2.1.3 for H_2CO) data products versions from the TROPOMI instrument on board the Sentinel-5 (S5p) precursor satellite. Only pixels satisfying the condition of quality value (qavalue > 0.75), within a 20km radius of the observation site were selected.

In order to reduce the uncertainties related to the impact of the apriori profile, we recalculated the TROPOMI tropospheric vertical columns using the median of the MAX-DOAS profile as the apriori profile. The equations giving rise to this transformation are described in equation 1 and 2. The MAX-DOAS profiles used are shown in Fig. 2, coupled with the AVK of the TROPOMI used to account for the accuracy of the satellite.

$$VCD_{MAX-DOAS}^{smoothed} = \sum_i AVK_i^{SP5} * C_i^{MAX-DOAS} \quad (1)$$

$$VCD_{SP5}^{recal} = VCD_{SP5} * \frac{VCD_{MAX-DOAS}}{VCD_{MAX-DOAS}^{smoothed}} \quad (2)$$

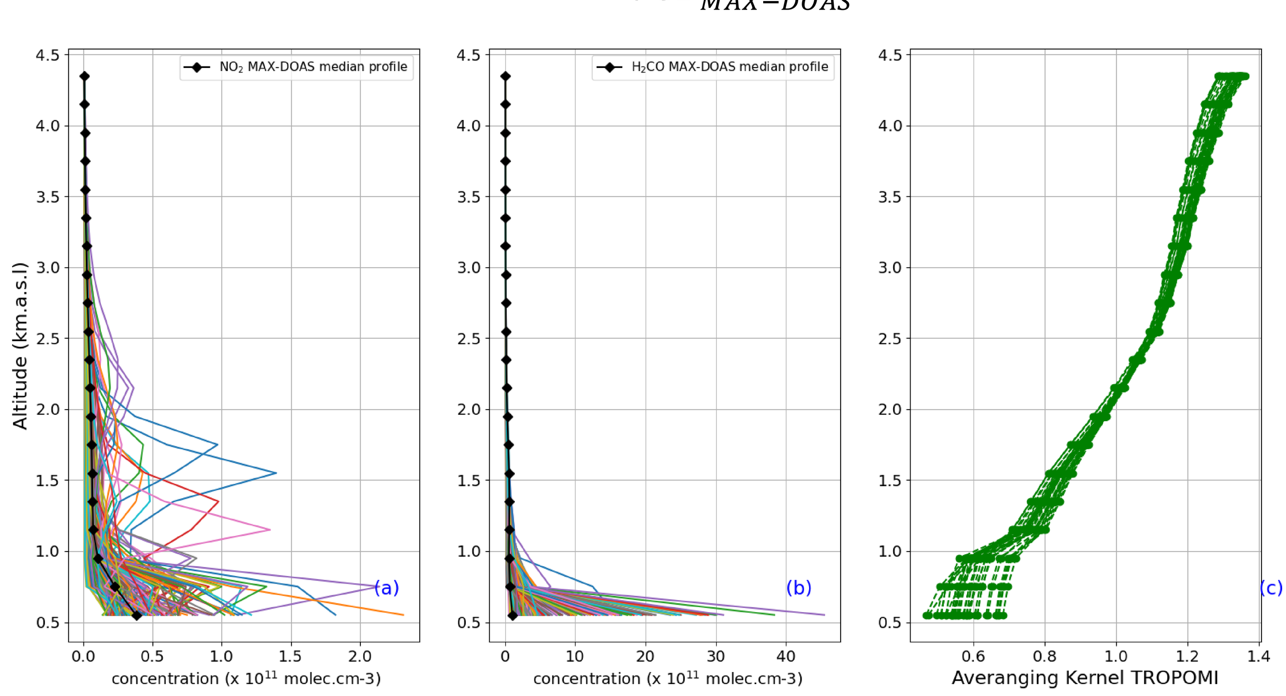

Figure 2. (panel a and b) : Sets of all MAX-DOAS profiles showing fluctuation over the year, associated with the median MAX-DOAS profile used to recalculate tropospheric columns (black dots) for NO_2 and H_2CO respectively. (panel c) : Example of NO_2 TROPOMI Averaging Kernels on June 13, 2020 showing a low sensitivity of TROPOMI in the first few kilometers.

RESULTS : Intercomparison of MAX-DOAS retrievals with TROPOMI data sets

Comparison between MAX–DOAS and TROPOMI NO_2

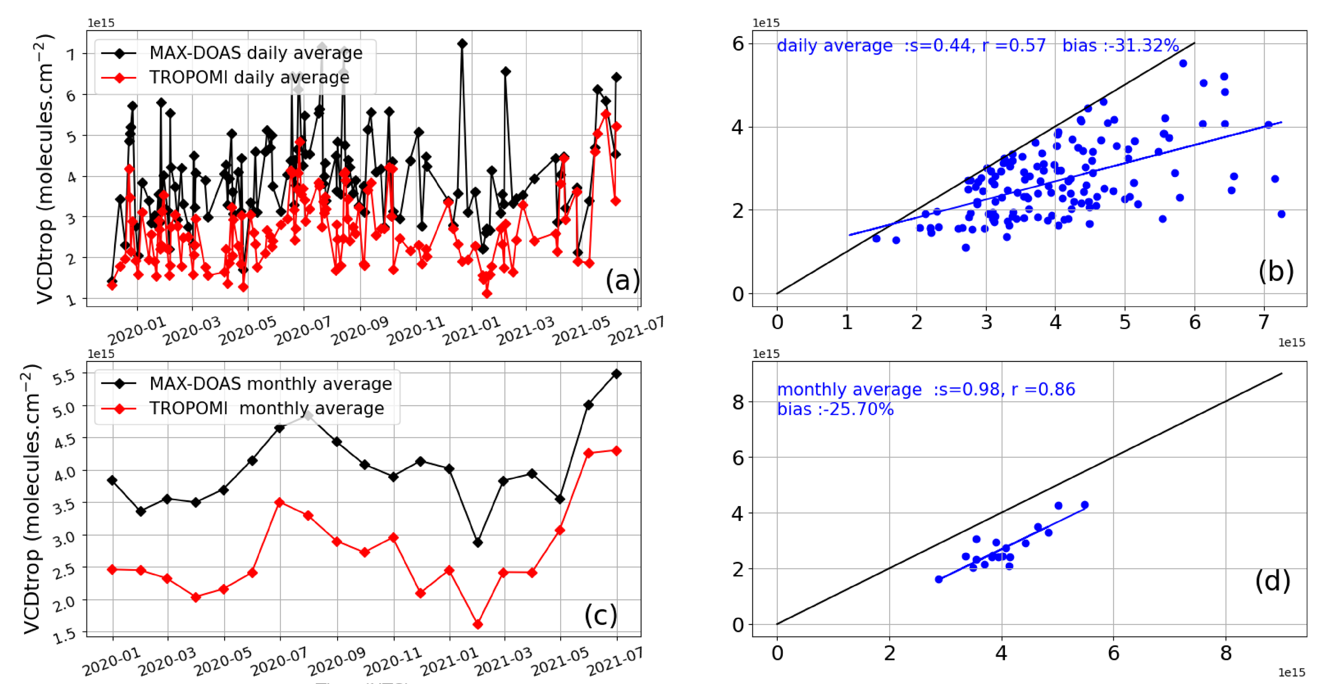

Figure 3. NO_2 time series of daily averages of MAX-DOAS (black dots) and TROPOMI (red dots). (panel b): linear regression between daily averages of MAX-DOAS and TROPOMI. (panel c): Time series of monthly averages of MAX-DOAS and TROPOMI. (panel d): linear regressions between the monthly means between the two datasets. The data cover the period from November 2019 to July 2021.

Comparison between MAX–DOAS and Tropomi H_2CO

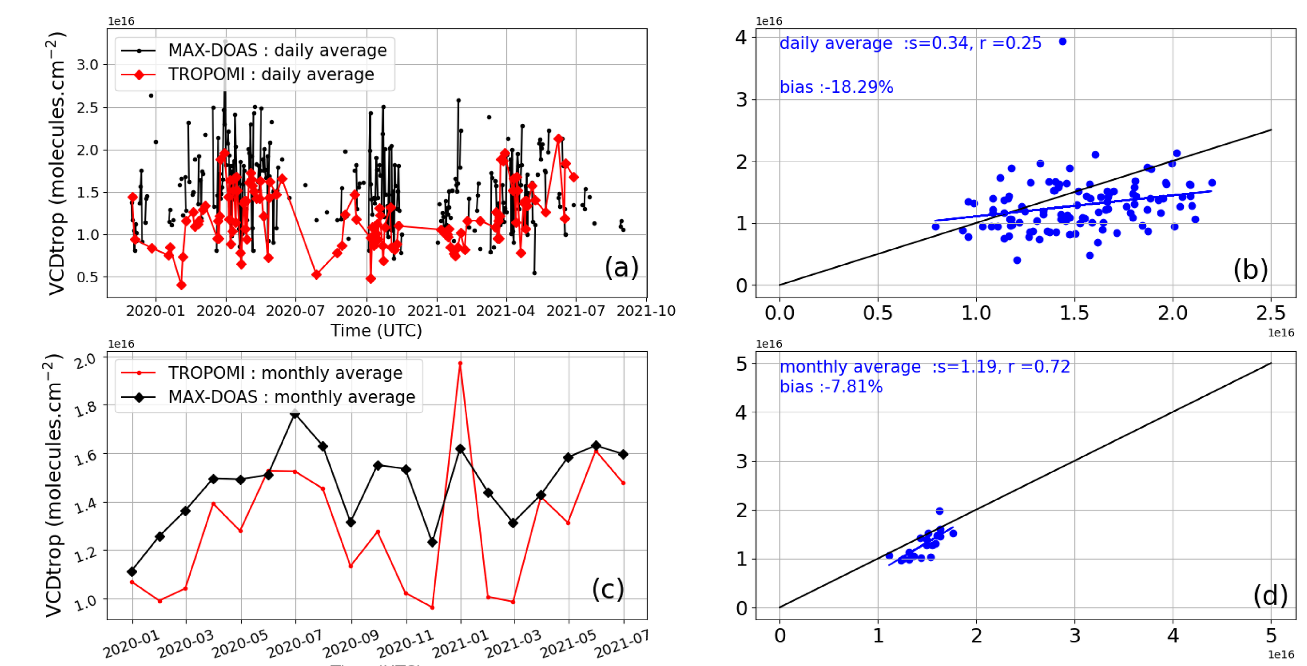

Figure 4. H_2CO time series of daily averages of MAX-DOAS (black dots) and TROPOMI (red dots). (panel b): linear regression between daily averages of MAX-DOAS and TROPOMI. (panel c): Time series of monthly averages of MAX-DOAS and TROPOMI. (panel d): linear regressions between the monthly means between the two datasets. The data cover the period from November 2019 to July 2021.

TROPOMI validation

The results of comparisons between MAX-DOAS and TROPOMI indicate a low correlation between the two datasets based on daily sampling. These results are greatly improved when we use sampling based on monthly averages. Despite the correction applied, based on the use of the MAX-DOAS profile as a priori in the production of the new TROPOMI product used, we still notice the underestimation of TROPOMI in relation to the ground observations. This underestimation may be due to other parameters described in Lorentz et al. (2017) and not addressed in this study, such as aerosols, surface albedo, metrological conditions. The city of Kinshasa is strongly dominated by aerosols and cloud cover, and these parameters may influence the current comparisons.

FUTURE PLANS

- Derivation and extraction of emissions that may have an influence on the observations made.
- Comparison of the top-down and bottom-up approach in understanding the observed bias between GEOS-Chem and MAX-DOAS.
- Comparison of the GEOS-Chem model with the MAGRITTE (**Model of Atmospheric composition at Global and Regional scales using Inversion Techniques for Trace gas Emissions**) model in order to evaluate the consistency of the observed negative bias.

Intercomparison of GEOS chem with TROPOMI and MAX-DOAS data sets

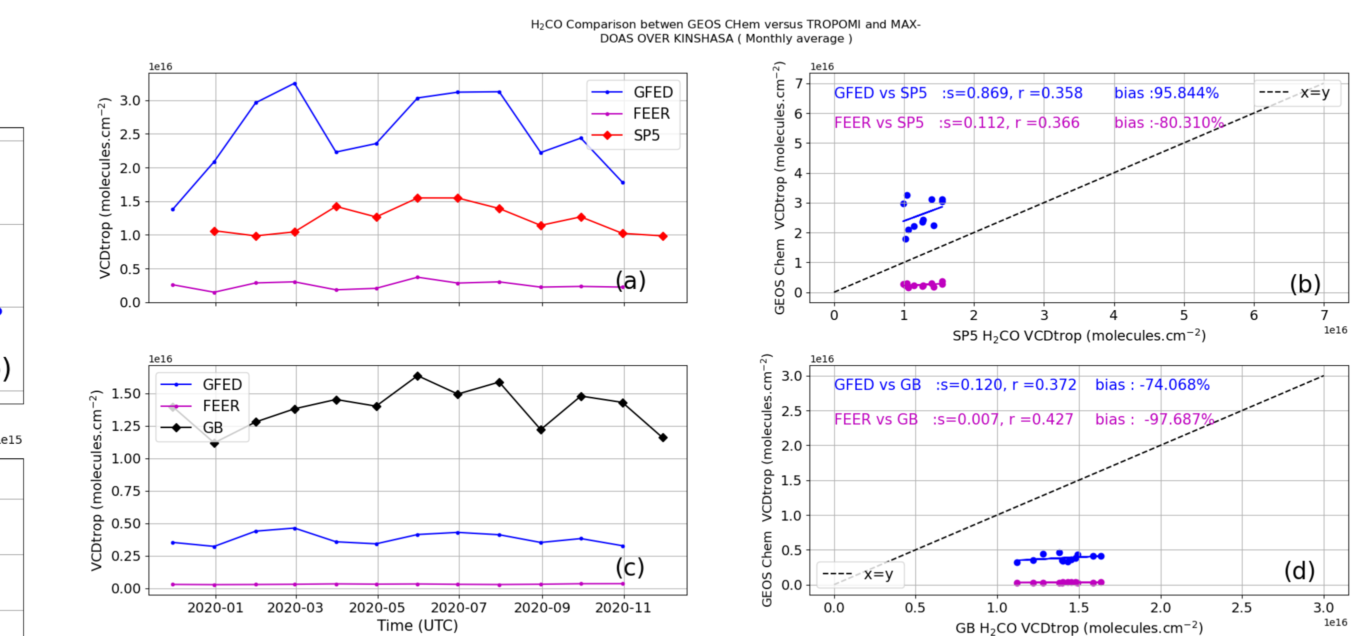

Figure 5. NO_2 time series of monthly means of GEOS-Chem simulations using the 2 biomass burning emission datasets GFED and FEER compared to TROPOMI (panel a) and MAX-DOAS (panel b) observations. Results from comparisons between the two datasets: TROPOMI vs GEOS-Chem (panel b), MAX-DOAS vs GEOS Chem (panel c).

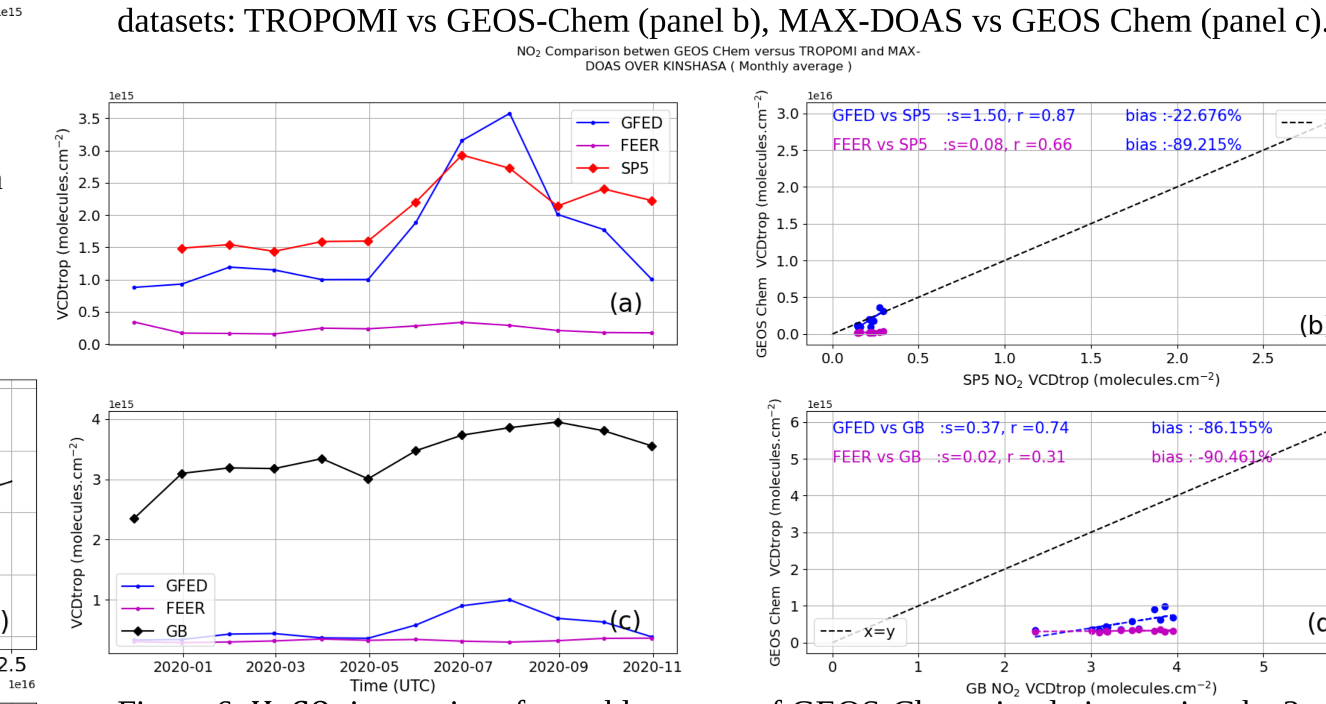

Figure 6. H_2CO time series of monthly means of GEOS-Chem simulations using the 2 biomass burning emission datasets GFED4 and FEER1.0 compared to TROPOMI (panel a) and MAX-DOAS (panel b) observations. Results from comparisons between the two datasets: TROPOMI vs GEOS-Chem (panel b), MAX-DOAS vs GEOS Chem (panel c).
FEER1.0 :Fire Energetics and Emissions Research version 1.0
GFED v4: Global Fire Emissions Database version 4.

GEOS Chem bias

We note an underestimation of the model compared to the ground observations. An approximate bias of -80% is observed between the two datasets for both molecules. This difference can be explained by several reasons related to the accuracy of GEOS-Chem which is generally affected by several factors such as its resolution, emission inventories, model chemistry involving the molecules studied (e.g., Marais et al. 2021). The emission inventories used in the model have a different and smaller resolution than the model. The different tests on biomass burning (BB) carried out in our study show how much the choice of BB could affect the results simulated by the model. And we note that the large contributions of emissions in all the inventories used in our simulations come from the BBs, especially those of GFED.

REFERENCES

1. Lorente, A., Boersma, K. F., Yu, H., Dörner, S., Hilboll, A., Richter, A., Liu, M., Lamsal, L. N., Barkley, M., Smedt, I. D., Roozendael, M. V., Wang, Y., Wagner, T., Beirle, S., Lin, J. T., Krotkov, N., Stammes, P., Wang, P., Eskes, H. J., and Krol, M.: Structural un-certainty in air mass factor calculation for NO2 and HCHO satellite retrievals, Atmospheric Measurement Techniques, 10, 759–782,https://doi.org/10.5194/amt-10-759-2017, 2017
2. Liousse, C., Assamoi, E., Criqui, P., Granier, C., and Rosset, R.: Explosive growth in African combustion emissions from 2005 to 2030, Environ. Res. Lett, 9, https://doi.org/10.1088/1748-9326/9/3/035003, 2014.
3. Marais, E. A., Roberts, J. F., Ryan, R. G., Eskes, H., Folkert Boersma, K., Choi, S., Joiner, J., Abuhassan, N., Redondas, A., Grutter, M., Cede, A., Gomez, L., & Navarro-Comas, M. (2021). New observations of NO2 in the upper troposphere from TROPOMI. *Atmospheric Measurement Techniques*, *14*(3), 2389–2408. https://doi.org/10.5194/amt-14-2389-2021
4. Ma, J. Z., Beirle, S., Jin, J. L., Shaiganfar, R., Yan, P., and Wagner, T.: Tropospheric NO2 vertical column densities over Beijing: Results of the first three years of ground-based MAX-DOAS measurements (2008-2011) and satellite validation, Atmospheric Chemistry and Physics, 13, 1547–1567, https://doi.org/10.5194/acp-13-1547-2013, 2013.
   Marais, E. A. and Wiedinmyer, C.: Air Quality Impact of Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa), Environmental Science and Technology, 50, 10 739–10 745, https://doi.org/10.1021/acs.est.6b02602, 2016.
5. McFarlane, C., Isevulambire, P. K., Lumbuenamo, R. S., Ndinga, A. M. E., Dhammapala, R., Jin, X., McNeill, V. F., Malings, C., Subramanian, R., & Westervelt, D. M. (2021). First measurements of ambient pm2.5 in kinshasa, democratic republic of congo and brazzaville, republic of congo using field-calibrated low-cost sensors. *Aerosol and Air Quality Research*, *21*(7). https://doi.org/10.4209/aaqr.200619
6. Yombo Phaka, R., Merlaud, A., Pinardi, G., Mahieu, E., Hendrick, F., Friedrich, M. M., Fayt, C., Van Roozendael, M., Djibi, B. L., Bopili Mbotia Lepiba, R., Phuku Phuati, E., and Mbungu Tsumbu, J.-P.: First Ground-Based Doas Measurements of No2 At Kinshasa and Comparisons With Satellite Observations, Journal of Atmospheric and Oceanic Technology, pp. 1291–1304, https://doi.org/10.1175/jtech-d-20-0195.1, 2021

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