Brazilian Atmospheric Inventories - BRAIN: A comprehensive database of air quality in Brazil

MS No.: essd-2023-305

Dear editor,

We greatly appreciate the reviewer comments and suggestions, which are very constructive and have contributed to enhance the content of the revised manuscript. Please find below the reply to all the reviewer comments.

Best regards

Leonardo Hoinaski and coauthors

Reply to comments of reviewer:

Comment #1: First of all, it is important for the authors to clarify the site locations. When

comparing the BRAIN with Artaxo et al., 2013 results.

Reply: We included new analyses comparing data from BRAIN and Sentinel-5P TROPOMI at the sites described in Martin et al., (2016) (background and urban areas). We adopted reanalysis and satellite data to compare the data from the same year as the BRAIN simulations, since GoAmazon campaigns are not available and Artaxo's work uses data from 2013.

We compared BRAIN and Sentinel-5P TROPOMI at T0t/TT34 (located at the ZF2 ecological reserve - low anthropogenic influence), T1 (Manaus - urban area), and T0a (upwind of Manaus – background area) sites as recommended by the reviewer. We used data from CO, NO_2, O_3 and PM_{10} from BRAIN and tropospheric column of CO, NO_2, and O_3 from Sentinel-5P TROPOMI.

We have included the following sentences in the manuscript:

To analyze BRAIN's performance in background regions (with low anthropogenic influence), we extracted data from two forested sites in the Brazilian North region. We used as reference the sampling sites of the GoAmazon experiment (Martin et al., 2016) named T0a (forested site situated 154.1 km from the Manaus urban area) and T0t/TT34 (a Broken canopy, forested site situated 60.9 km from the Manaus). Sentinel-5P TROPOMI data spatially aligned to BRAIN resolution was also extracted for comparison. A buffer of 0.2° around the sites selected the data of CO, O_3, and NO_2 from both datasets. The results revealed that BRAIN captured the seasonal profile at T0a (Figure 11), showing a moderate correlation with tropospheric column measurements of Sentinel-5P TROPOMI, especially for CO and O_3. BRAIN estimates are consistent with concentrations in background areas of CO (up to ~ 350 ppb), O_3 (up to ~ 0.04 ppm), and NO_2 (up to ~ 2.5 $\mu g.m^{-3}$) at T0t/TT34 (Figure 12) and T0a. However, it was not able to capture the seasonal profile at T0t/TT34.

We also analyzed BRAIN results in the Manaus urban area. We adopted the sampling site of the GoAmazon experiment (Martin et al., 2016) named T1 (INPA campus in Manaus). Compared with Sentinel-5P TROPOMI data, BRAIN fairly reproduced the temporal pattern of CO, O_3, and NO_2 in the T1 site (Figure 13).

Comment #2: Manaus is not a background region. During the GoAmazon 204/5 experiment (Martin et al., 2016) many different sites were used to understand how the interaction between biogenic and urban emissions can affect the aerosol life cycle and its impacts on the cloud, radiation and water cycle in the Amazon region. To investigate background regions during the GoAmazon experiment the authors should look at sites: T0a (ATTO), T0z (TT34-ZF2), T0e. Those sites were upwind of Manaus, which means almost no influence from Manaus anthropogenic emission. T1 site (located in Manaus) is the site that was representing the gas/particle distribution in the city.

Reply: Thanks for the suggestions. We compared data of BRAIN and Sentinel-5P TROPOMI during 2019 at the different sites as suggested. To analyze BRAIN results in background regions, we adopted a forested site upwind of Manaus (named T0a in the GoAmazon reference), situated 154.1 km from the Manaus urban area. We also analyzed BRAIN results in a Broken canopy, forested site north of Manaus (named T0t/TT34 in the GoAmazon reference), situated 60.1 km from the Manaus urban area. We also analyzed BRAIN results in the Manaus urban area (T1 site). Our analysis revealed that BRAIN clearly captured the seasonal patterns in T1 and T0a, especially for CO and O_3 concentrations. BRAIN estimates are consistent with concentrations in background areas of CO (up to ~ 350 ppb) and NO_2 (up to ~ 2.5 $\mu g.m^{-3}$) at T0t/TT34 (Figure 12) and T0a. However, it was not able to capture the seasonal profile at T0t/TT34. It shows that BRAIN could be an interesting alternative to fill the data gaps in background areas.

We have included the following sentences and figures in the manuscript:

> *To analyze BRAIN's performance in background regions (with low anthropogenic influence), we extracted data from two forested sites in the Brazilian North region. We used as reference the sampling sites of the GoAmazon experiment (Martin et al., 2016) named T0a (forested site situated 154.1 km from the Manaus urban area) and T0t/TT34 (a Broken canopy, forested site situated 60.9 km from the Manaus). Sentinel-5P TROPOMI data spatially aligned to BRAIN resolution was also extracted for comparison. A buffer of 0.2° around the sites selected the data of CO, O_3, and NO_2 from both datasets. The results revealed that BRAIN captured the seasonal*

profile at T0a (Figure 11), showing a moderate correlation with tropospheric column measurements of Sentinel-5P TROPOMI, especially for CO and O_3. BRAIN estimates are consistent with concentrations in background areas of CO (up to ~ 350 ppb), O_3 (up to ~ 0.04 ppm), and NO_2 (up to ~ 2.5 $\mu g.m^{-3}$) at T0t/TT34 (Figure 12) and T0a. However, it was not able to capture the seasonal profile at T0t/TT34.

We also analyzed BRAIN results in the Manaus urban area. We adopted the sampling site of the GoAmazon experiment (Martin et al., 2016) named T1 (INPA campus in Manaus). Compared with Sentinel-5P TROPOMI data, BRAIN fairly reproduced the temporal pattern of CO, O_3, and NO_2 in the T1 site (Figure 13).

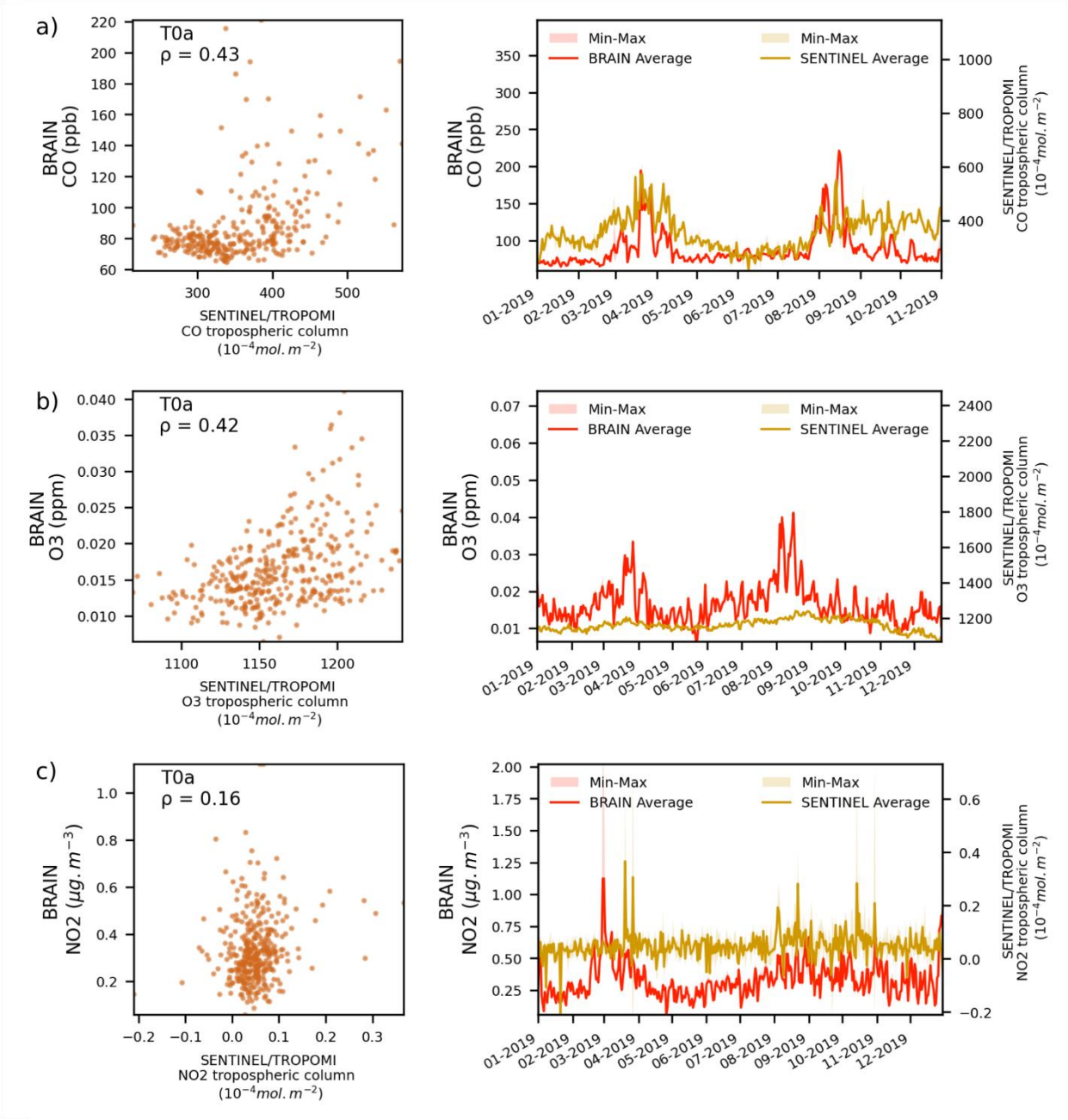

Figure 11. Scatterplot and daily time series of CO (a), O_3 (b), and NO_2 (c) from BRAIN and Sentinel-5P TROPOMI at T0a (GoAmazon reference). Values extracted using a buffer of 0.2° around the station.

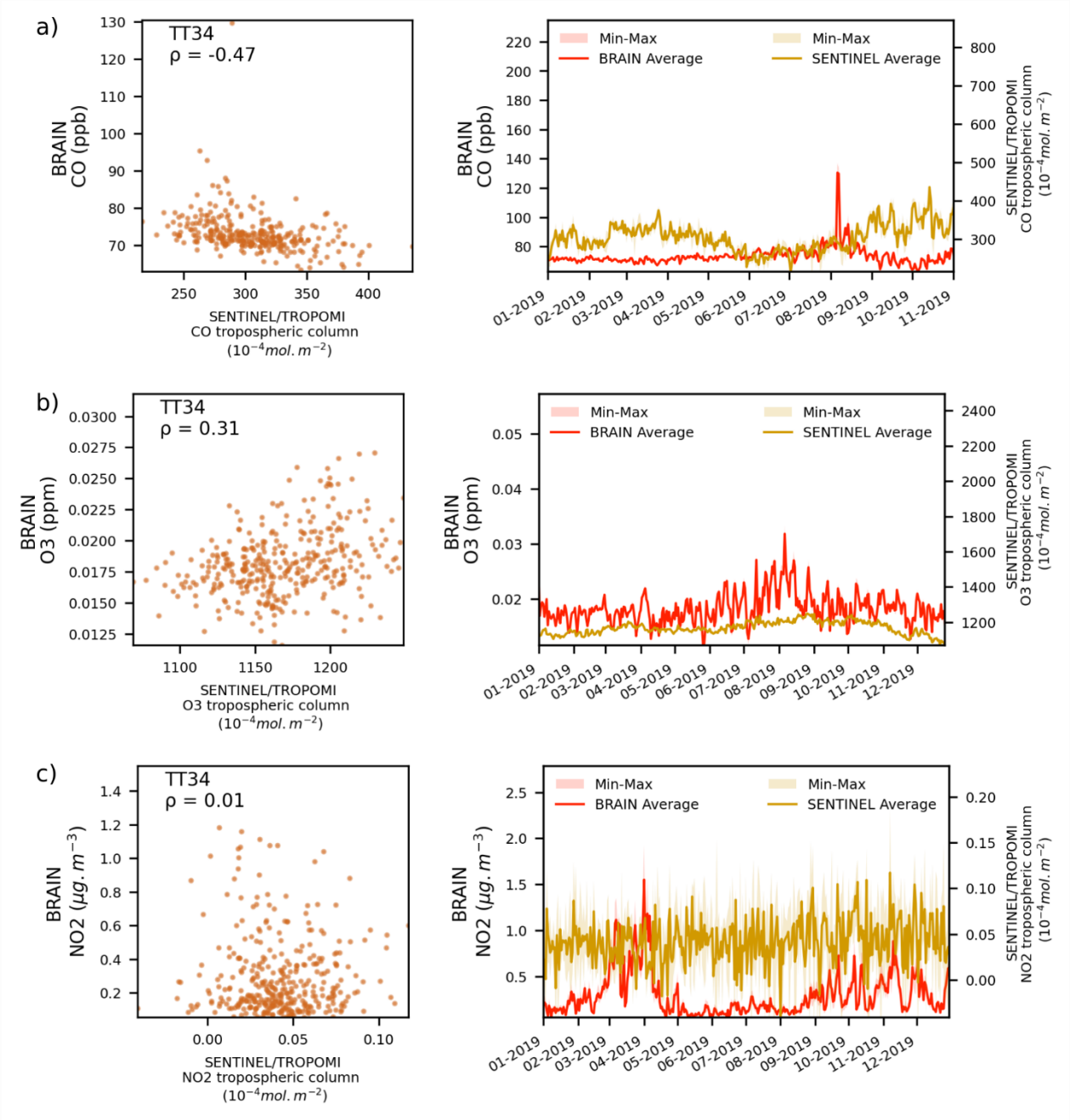

Figure 12. Scatterplot and daily time series of CO, O_3, and NO_2 from BRAIN and Sentinel-5P TROPOMI at T0t/TT34 (GoAmazon reference). Values extracted using a buffer of 0.2° around the station.

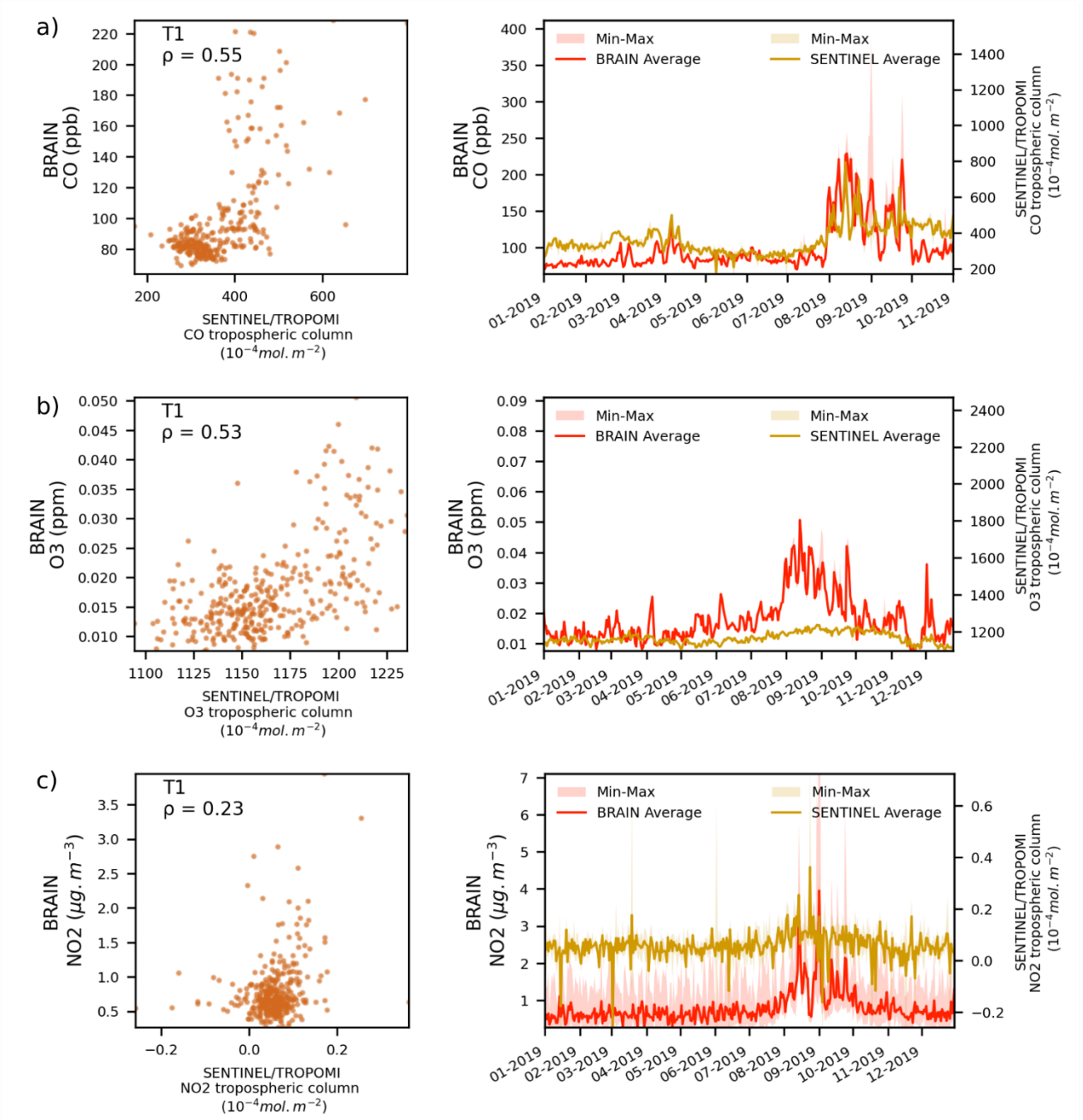

Figure 13. Scatterplot and daily time series of CO (a), O_3 (b), and NO_2 (c) from BRAIN and Sentinel-5P TROPOMI at T1 (GoAmazon reference). Values extracted using a buffer of 0.2° around the station.

We also removed the "*background areas such as Manaus in Amazonas*," from the manuscript.

Comment #3: In the Figure SM8, are the authors spatially averaging in the Manaus city? It is very hard to know the values on the dark red line (average). Can the authors provide a more

understandable figure?

Reply: Yes, the lineplot in Figure SM8 represents the spatial average and minimum-maximum of pixels within Manaus. The idea was providing a figure in each Brazilian capital. We provided a new Supplementary Material (SM19) only with averages of BRAIN data to improve the reader's understanding.

Comment #4: For the site TT34 located at the ZF2 ecological reserve (situated 60 km to the north-northwest of Manaus urban area in the Cuieiras Biological Reserve), based on the figure provided, BRAIN is not showing seasonal differences on the PM2.5 values. This site is located in a region with low anthropogenic influence. Why are the authors not comparing BRAIN outputs sampled at the forest site TT34 location?

Reply: We provided a new analysis comparing BRAIN and *Sentinel-5P TROPOMI* at TT34. Our analysis revealed that BRAIN results at TT34 are consistent with background concentrations. However, we haven't found meaningful correlation at TT34, which could be explained by the lower influence of anthropogenic sources.

We have included the following sentences and figures in the manuscript:

> *To analyze BRAIN's performance in background regions (with low anthropogenic influence), we extracted data from two forested sites in the Brazilian North region. We used as reference the sampling sites of the GoAmazon experiment (Martin et al., 2016) named T0a (forested site situated 154.1 km from the Manaus urban area) and T0t/TT34 (a Broken canopy, forested site situated 60.9 km from the Manaus). Sentinel-5P TROPOMI data spatially aligned to BRAIN resolution was also extracted for comparison. A buffer of 0.2° around the sites selected the data of CO, O_3, and NO_2 from both datasets. The results revealed that BRAIN captured the seasonal profile at T0a (Figure 11), showing a moderate correlation with tropospheric column measurements of Sentinel-5P TROPOMI, especially for CO and O_3. BRAIN estimates are consistent with concentrations in background areas of CO (up to ~ 350 ppb), O_3 (up to ~ 0.04 ppm), and NO_2 (up to ~ 2.5 $\mu g.m^{-3}$) at T0t/TT34 (Figure 12) and T0a. However, it was not able*

to capture the seasonal profile at T0t/TT34.

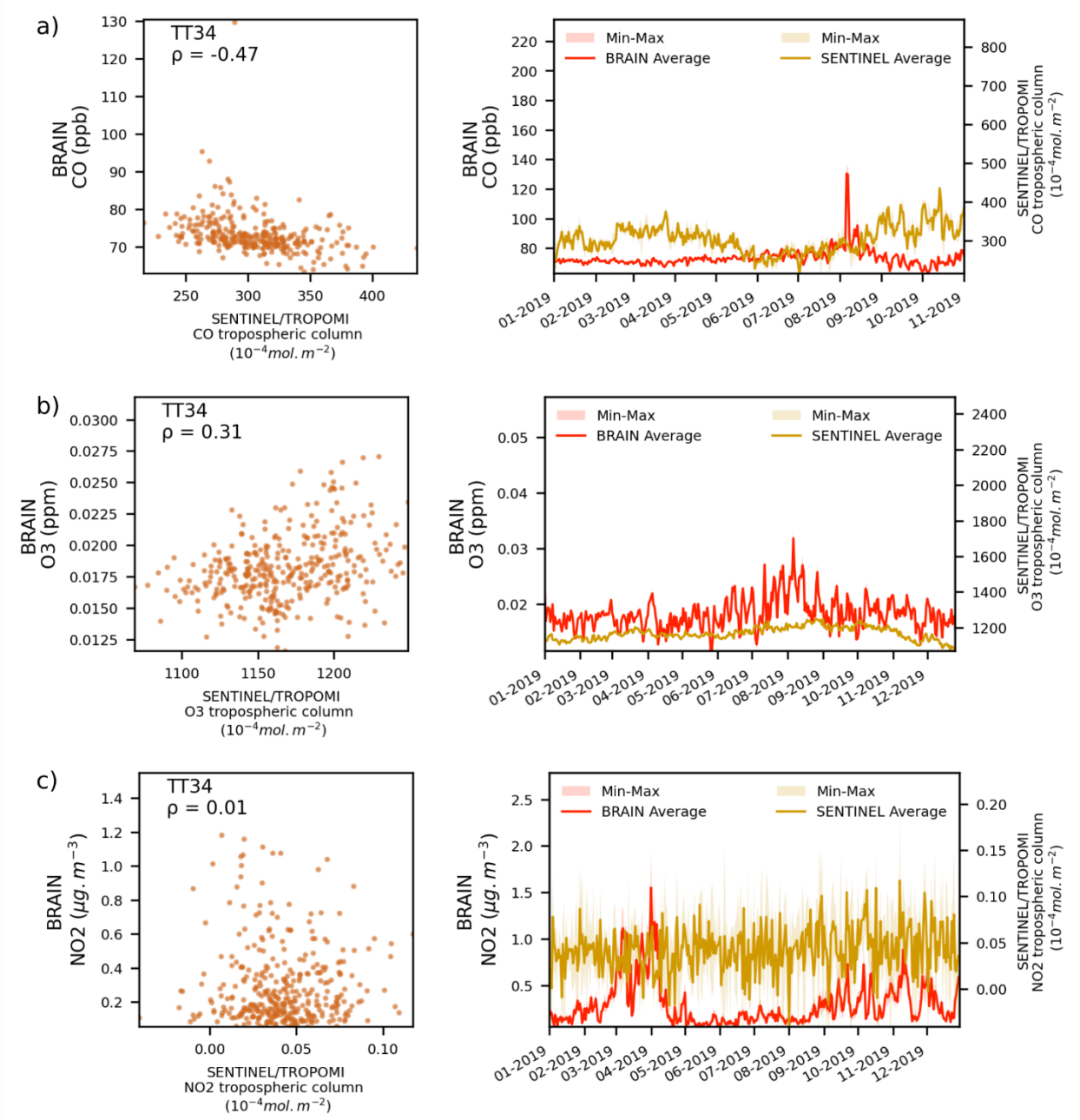

Figure 12. Scatterplot and daily time series of CO, O_3, and NO_2 from BRAIN and Sentinel-5P TROPOMI at T0t/TT34 (GoAmazon reference). Values extracted using a buffer of 0.2° around the station.

Comment #5: About the site in Porto Velho (PVH was located near Porto Velho city). The authors

should follow the same approach, compare the BRAIN results with the PVH site location, instead of spatially averaging a large area.

Reply: Ok, we now have compared with site locations at T0a, TT34, and T1. We will keep the spatially averaged figure in the manuscript since it brings a comparison between the databases (MERRA2, Sentinel-5P TROPOMI, and BRAIN).

Comment #6: Related with the PM2.5 analysis: The authors compared Artaxo et al., 2013 figure PM10 time series with BRAIN's PM2.5 time series in Porto Velho (Figure SM8). The seasonal change, with a peak in the dry season is shown in the BRAIN. Although it is important to emphasize the difference on the aerosol mode (particle size). Artaxo et al., shows fine and corse mode. BRAIN shows PM2.5 (fine particles) which is more similar with the coarse mode of the time series showed in Artaxo et al., 2013.

Reply: Ok. We included a new sentence in the discussion of the revised manuscript highlighting similarities with the coarse mode of the time series shown in Artaxo et al., 2013.

We have included the following sentences and figures in the manuscript:

> *However, BRAIN $PM_{2.5}$ estimates are closer to the coarse mode of the time series, rather than the fine mode shown in Artaxo et al., (2013).*

Comment #7: Another point is related with the O3 analysis. BRAIN captured the absolute magnitude of O3 in September 2019 when compared with observed averaged data between 2009 to 2012 from Artaxo et al., 2013. It is important to mention the overestimation between the samples. BRAIN shows O3 2.7 times higher that the observation data (dry season). And a factor of 2 higher for the wet season of O3 values.

Reply: Ok. We mentioned the overestimation of BRAIN concerning the Artaxo et al., (2013) results. We also explain that Artaxo's campaing was conducted in 2013, while BRAIN in 2019.
We have rephrase the sentence in the manuscript:

BRAIN captures seasonal patterns and the absolute magnitude of $PM_{2.5}$ in the Northwest of the Amazonas state (near the Amazon Tall Tower Observatory -ATTO) presented by Artaxo et al., (2013). It shows that our database can reproduce the concentrations in background areas (far from highly urbanized centers). Comparing BRAIN with observations at heavy biomass burning impacted sites in south-western Amazonia (Porto Velho) (Artaxo et al., 2013) revealed that BRAIN can capture seasonal variations caused by wet and dry seasons and the magnitude of average and peak concentrations. However, BRAIN estimates are closer to the coarse mode of the time series, rather than the fine mode shown in Artaxo et al., 2013. Even though BRAIN has captured the O_3 pattern observed by Artaxo et al. (2013), the estimates are around 2.7 higher than the observations in the dry season and a factor of 2 higher for the wet season. It is worth mentioning that BRAIN uses 2019 data, while Artaxo et al. (2013) consists of a sampling campaign from 2008 to 2012.

Comment #8: Figure 8 (Annual average concentration of CO and NO2 from BRAIN original resolution (a, c), SENTINEL/TROPOMI regridded to BRAIN resolution (b, d)). Can the authors convert the data to the same unit. Ex: all in PPB? In the same Figure, why is that so much of CO in the ocean region - b) Regrided SENTINEL/TROPOMI?

Reply: *Sentinel-5P TROPOMI* data are provided in ($mol.m^{-2}$) unit, representing the density of pollutants in the tropospheric column. For this reason, *Sentinel-5P TROPOMI* data cannot be straight-forward converted to surface concentration.

Comment #9: The idea of having a dataset as BRAIN is very needed specially due to the current lack of numerical simulated dataset available to work with air quality in Brazil. Also because not everyone that works with meteorological and/or air quality knows how to produce and work with atmospheric models. Nevertheless, the first step on evaluating and validating a modeling dataset in order to analyze regions beyond the AQS/aircraft is by comparing the modeling dataset against the most observational data available. After that, the modeling data set can be used to allows us to directly assess the regions where AQS are not available. I would expect most of the analysis in this paper to occur in areas with high AQS.

Reply: We appreciate the compliments.

Comment #10: Question: Which chem_opt option are you using in the namelist.input file?

Reply: If you mean chemical option of chemical transport model, we used Carbon Bond 06 mechanism. We provide all details of our simulation in the supplementary materials, including WRF namelists and CMAQ scripts.

References:

Martin, S.T., Artaxo, P., Machado, L.A.T., Manzi, A.O., Souza, R.A.F., Schumacher, C., Wang, J., Andreae, M.O., Barbosa, H.M.J., Fan, J., Fisch, G., Goldstein, A.H., Guenther, A., Jimenez, J.L., Pöschl, U., Silva Dias, M.A., Smith, J.N., and Wendisch, M. (2016). Introduction: Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5). Atmos. Chem. Phys., 16, 4785–4797, https://doi.org/10.5194/acp-16-4785-2016