

author comments on the manuscript “Monitoring CO emissions of the metropolis Mexico City using TROPOMI CO observations”, reviewer 1

We would like to thank the reviewer for the constructive comments that aided us to improve our manuscript. In this document we provide our replies to the reviewer’s comments. The original comments made by the reviewer are numbered and typeset in italic and bold face font. Following every comment, we give our reply. Here line numbers, page numbers and figure numbers refer to the original version of the manuscript, if not stated differently. Additionally, the revised version of the manuscript is added.

1 Major Comments

1. *One of the main concerns is regarding the CO background concentration and chemistry. Authors assume a time invariant CO background concentration, while I believe background processes in the region of interest and its surrounding are quite important. I highly suggest to describe in detail why a constant CO background has been used. Please explain in detail how the background CO flowing into the domain produced by all non-metropolis Mexico City (10 districts) sources, including, non-metropolis Mexico City fires, is treated. Considering the relatively long lifetime of CO transport is extremely important.*

adjusted This is a misunderstanding, we are not assuming a time invariant CO background concentration in our study. Figure 2a shows for each TROPOMI overpass of Mexico City which background CO concentration was used. The time series clearly reflects elevated background CO concentrations during dry season (e.g. fire contribution) and low background CO during rainy season over Mexico City. Actually, we found, that the CO background concentration is a crucial component of our emission inversion scheme and therefore, decided to retrieve it together with the CO emission of the 10 city districts of Mexico (parameters $\alpha_{bg}, \alpha_{elev}$ in Equation 6). This ensures that the inversion scheme has the capability to decided itself which part of the TROPOMI measurement to interpreted as background and which as contribution from the city districts. Hence, all contribution from other sources excluding the 10 city districts will be represented by the fitted background parameters.

To make this clearer, we changed the definition of the forward model in section 3.1. Now the background parameters are exclusively introduced as effective fit parameters in Eq. 5. Furthermore, we changed the paragraph p5, l27 from:

“ Finally, to improve the capability of the forward model to fit TROPOMI observations, we induce a linear altitude dependence of the simulated CO column $\mathbf{k}_{elv} = z - z_{ref}$. Here, z is the mean elevation in the TROPOMI CO ground pixels and $z_{ref} = 2240$ m the reference altitude which is set to the elevation of Mexico City.

$$\mathbf{F}_{sat}(E_1, \dots, E_{10}, \alpha_{bg}) = \sum_{i=1}^{10} \mathcal{O}(\tilde{\mathbf{k}}_i) E_i + \mathbf{k}_{bg} \alpha_{bg} + \mathbf{k}_{elv} \alpha_{elv} \quad (5)$$

With these additional degrees of freedom the forward model can mitigate shortcomings of the WRF simulations using a spatially constant CO background. ”

to

“ To improve the capability of our forward model to fit TROPOMI observations, we introduce a spatially constant CO background field \mathbf{k}_{bg} and an altitude dependence term $\mathbf{k}_{elv} = z - z_{ref}$ with corresponding scaling factors α_{bg} and α_{elv} . Here, z is the respective elevation of the TROPOMI CO ground pixels and $z_{ref} = 2240$ m is an arbitrary reference altitude set to the elevation of Mexico City,

$$\mathbf{F}_{sat}(E_1, \dots, E_{10}, \alpha_{bg}, \alpha_{elv}) = \sum_{i=1}^{10} \mathcal{O}(\tilde{\mathbf{k}}_i) E_i + \mathbf{k}_{bg} \alpha_{bg} + \mathbf{k}_{elv} \alpha_{elv} . \quad (5)$$

”

2. *Furthermore, biogenic non-methane VOCs emitted from vegetation might be important as a source for the chemical production of CO in the atmosphere. In the manuscript, I did not find information regarding these contribution, maybe it is too small for the metropolis?, what about the transport of the surroundings to the districts. It would be important to add a description on this.*

adjusted The WRF model in this study is run in transport only mode. Hence, the chemical production of CO is not accounted for. However, it should be small compared to the other sources. All type of contributions like this (biogenic) or from outside the domain such as fires, power plants, other cities as well as contribution from global CO is compensated by the fitted background parameters $\alpha_{bg}, \alpha_{elev}$. Hence, when the background CO field becomes too complex or inhomogeneous e.g. as discussed for the CO from wild fires in Fig. 3, our approach will fail to reproduce the TROPOMI measurements and these cases are rejected.

We changed the sentence p4,114 from:

“Further, the forward model assumes linear dependence of CO background field \mathbf{k}_{bg} with scaling parameter α_{bg} ...”

to

“These two effective model components account for CO contribution over the Mexico City area originating from outside the model domain such as fires, power plants, biogenic production, other cities as well as the long-range transport (Borsdorff et al., 2019) and an altitude dependent linear vertical gradient of the CO columns. Both do not interfere with any localized emission sources. They mitigate shortcomings of the WRF-chem simulations ignoring CO boundary conditions at the model domain. ”

3. *Lastly, according to the authors the configuration of the model does not account for atmospheric chemistry, does that mean that Gas-phase Chemistry is not included?. Similarly, please include a description of why this configuration was chosen.*

adjusted

We changed the sentence p3,129 from:

“...and does not account for atmospheric chemistry (Dekker et al., 2017).”

to

“ We ignore photo-chemical oxidation and secondary production of CO in the atmosphere (chem_opt option 106 (RADM2-KPP), as a tracer with gaschem off), which is justified by the long lifetime of CO compared with the size of the model domain as discussed by Dekker et al. (2017). Especially, for the region of Mexico City the contribution of atmospheric chemistry to the total CO concentration is less than 3% as presented by Mejia (2020). Hence, WRF-chem simulates the transport of CO surface emission as traces as done by e.g. Borsdorff et al. (2019), Dekker et al. (2017, 2018). ”

2 Specific Comments

1. *Authors recognize the possible error sources, and if I understand correctly authors estimate uncertainties in the inversion, I highly suggest to include the uncertainties of emissions in the abstract.*

adjusted

We changed the sentence p1,18 from:

“...0.10 Tg/yr and 0.08 Tg/yr CO”

to

“ 0.10 ± 0.004 Tg/yr and 0.09 ± 0.005 Tg/yr CO ”

We changed the sentence p1,110 from:

“ For CDMX, TROPOMI estimates emissions of 0.14 Tg/yr ...”

to

“ On the other hand for Ciudad de Mexico, TROPOMI estimates emissions of 0.14 ± 0.006 Tg/yr CO, ...”

We changed the sentence p1,111 from:

“ ACDMX area, however, has a higher emissions with 0.29 Tg/yr according to TROPOMI observations ...”

to

“ ...Arena Ciudad de Mexico the emission is 0.28 ± 0.01 Tg/yr according to TROPOMI observations ...”

We changed the sentence p1,110 from:

“ ... (0.43 Tg/yr TROPOMI versus 0.39 Tg/yr adapted INEM emissions). ”

to

“ ... (0.42 ± 0.016 Tg/yr TROPOMI versus 0.39 Tg/yr adapted INEM emissions).”

In addition we changed the same statements in the results and conclusion section p8,l30-43.

2. *P1, L2. It is mentioned that 551 overpasses are analyzed, please specify the exact time period. The season(s) might be relevant.*

adjusted

We changed the sentence p1,l2 from:

“... (more than 2 years of measurements) using ...”

to

“... we analyze TROPOMI observations over Mexico City in the period 14 November 2017 to 25 August 2019 by ...”

3. *P1, L4. It is not clear to me if you use WRF coupled with Chemistry (WRF-Chem)?*

adjusted

We changed the sentence p1,l4 from:

“... regional Weather Research and Forecasting (WRF) model to conclude ...”

to

“... regional Weather Research and Forecasting (WRF-chem) model to conclude ...”

Accordingly, we changed the Acronym in the whole document

4. *P1, L8. Do you identify the sources missing in the INEM in Tula and Pachuca?*

adjusted

We added the following sentence p8,l30:

“It is not yet clear what sources are missing in the inventory, this needs to be addressed in future studies. However, we identified an oil refinery and a power plant near to Tula and cement and lime kilns near to Pachuca that could contribute to the CO emissions. ”

5. *P1, 14. It is mentioned: "CDMX and ACDMX follow a clear weakly cycle with a minimum during the weekend" does this mean that the weekend effect is not found in the other regions?*

not adjusted

No, but we cannot not conclude on it yet. We need to wait for more TROPOMI data to analyze the remaining districts.

6. *Section2, TROPOMI CO data set: In the current manuscript, I do not find a real value of including the FTIR observations, however it might be good to include it in this section. I suggest to include comparisons between TROPOMI and FTIR for coincident dates, do they compare ok?*

not adjusted The agreement between TROPOMI and the FTIR measurements is already analyzed in Borsdorff et al. (2018). We found in general a good agreement with a low bias. The FTIR measurements show that the weekly cycle in CO can be detected in the total column concentration and by that adds extra information to weekly cycle that is detected by in-situ measurements at the surface. Hence, we would like to keep the FTIR measurements here.

7. *Section 3.1, The WRF model. Important chemical parameterizations in the model are missing, e.g., what biogenic and biomass burning emissions are used?. What kind of boundary conditions?. Is the inflow of CO emitted by fires outside the region of interest included?. What time step is used?*

adjusted

We added the following sentence p3,l29:

“... (chem_opt option 106 (RADM2-KPP), as a tracer with gaschem off) ...”

Please also see major comment 3 of referee 1.

8. *P4, L17. Do you mean equation 1?*

corrected

9. *P5, L3-7. As in my major comment, it is a big assumption that local enhancements of CO are due to emissions of the city districts of the same day with a constant CO background?. It is well known that biomass/fire emissions can contribute significantly to the CO in the region. I wonder why an inflow of background CO is not taken into account, my understanding is that WRF-Chem can handle this.*

adjusted

Please also see our answer to the major comment of the referee. We changed the paragraph p5,l3-7 from: “ In our simulation of TROPOMI CO observations, we assume that the local enhancements of CO are due to emissions of the city districts of the same day, whereas emissions from outside the domain as well as the temporal accumulation of CO emission of the domain is described by the background CO field. Therefore, it means that the inferred emissions E_i represents an emission estimate of the urban district for the particular observation day. Moreover, the effective model parameter α_{bg} and α_{elv} may vary between different TROPOMI overpasses.

to

“ Finally, for the interpretation of our CO forward simulations, we make an important assumption. Although the WRF simulations account for the temporal accumulation of the localized CO emission over days and weeks, we allocate an emission estimate of the corresponding overpass time to each TROPOMI overpass. Here, we assume that a TROPOMI CO image is dominated by the emissions of the urban districts for the particular observation day, where the temporal accumulation of CO from previous days is partly described by the WRF simulation due to the corresponding scaling of the inventory and partly mitigated by fitting the nuisance parameter α_{bg} and α_{elv} . ”

10. *In order to have a sense of the spatial distribution of CO, I highly suggest to include the urban districts in Fig. 3.*

adjusted The figure is updated as suggested.

11. *P8, L2-9. It is not clear how the background concentration was estimated.*

adjusted

We changed the sentence p8,l1 from:

“ Fig. 2 shows the fitted CO background concentration and its annual cycle.”

to

“ Fig. 2 shows the CO background that was fitted as an auxiliary parameter during the inversion described in Sec. 3.2. The concentration and its annual cycle is shown. ”

12. *P8, L9. It is mentioned that the fire season many data cannot be considered, how many days (or percent) are excluded based on this?*

adjusted

We changed the sentence p8,l9 from:

“ Only fitting a scaling to a constant background field is not sufficient in this extreme cases and so during the fire season many data cannot be considered. . . .”

to

“ Only fitting a scaling to a constant background field is not sufficient in these extreme cases and so during the fire season many data cannot be considered (we excluded the month May and June 2019). ”

13. *Figure 4, It is hard to identify the districts on this figure, maybe you could include the contour/shapes of the districts.*

adjusted

The figure is updated as suggested.

14. *Figure 5. I recommend to follow the names of the districts as in Figure 1. Especially for Ciudad de Mexico and CDMX.*

adjusted

We updated Fig. 5,6,7, and 8 as well as the whole text of the Manuscript. The term “Ciudad de Mexico” is replaced by ACdMx and “CDMX” by “CdMx”. Hence, we are now following the nomenclature shown in Figure 1.

15. *Figure 5. why does Tulancingo have a zero emission?*

adjusted

We changed the sentence p9,111- from:

“ Moreover, the averaging kernel shows that the Final-fit inversion is insensitive to deviations of the Tulancingo emission from the prior estimate. Whereas the Pre-fit inversion estimates very small emissions for this district, the subsequent regularization changes the emission only marginally. ”

to

“ The figure shows that generally the averaging kernels have high values on the diagonal indicating high sensitivity to the quantity to be retrieved. It indicates that TROPOMI measurements can be used to distinguish emissions of the different urban districts of Mexico, with the exception of the emissions of district Tulancingo. Due to the small mean emission, the averaging kernel indicates a low sensitive of the data product. ”

16. *Figure 5. Is the number of collocations the same as the number of days?*

adjusted

We added the following sentence to Figure 5:

“ Here, a collocation corresponds to a specific day because TROPOMI overpasses the region only once. ”

17. *Figure 7. what does negative CO emission mean?*

adjusted

We added the following sentence p9,125:

“ The scatter of the data is still high and even includes negative values. Even though negative emissions are not physical we need to keep them in our analyzes because filtering negative noise can induce a positive bias in the mean. ”

18. *Figure 7. It is hard to believe that emissions on Sat and Sunday are very similar, what time does it represent the emissions?*

adjusted

We added the following sentence p9,129:

“ We found that the CO values on Saturday and Sunday are equally low. An explanation for this could be that the main source of CO in Mexico City during the week is traffic which is responsible for the weekly cycle and the remaining sources like cooking, water heating, etc. should not change much during the weekend. ”

19. *Figure 8. The weekly cycle of CO is considerably different than the weekly cycle of the emissions from Fig 7 (c), maybe I miss it but do you explain why?. Also, error bars from FTIR are extremely low, I do not think a standard deviation from the mean is the best way to characterize variability.*

not adjusted

We discussed this point on p9, 134. The variability of the weekly cycle is to high to conclude on its form yet. This will be revisited when we have more TROPOMI CO data available.

References

- Borsdorff, T., aan de Brugh, J., Hu, H., Hasekamp, O., Sussmann, R., Rettinger, M., Hase, F., Gross, J., Schneider, M., Garcia, O., Stremme, W., Grutter, M., Feist, D. G., Arnold, S. G., De Mazière, M., Kumar Sha, M., Pollard, D. F., Kiel, M., Roehl, C., Wennberg, P. O., Toon, G. C., and Landgraf, J.: Mapping carbon monoxide pollution from space down to city scales with daily global coverage, *Atmospheric Measurement Techniques Discussions*, 2018, 1–19, <https://doi.org/10.5194/amt-2018-132>, URL <https://www.atmos-meas-tech-discuss.net/amt-2018-132/>, 2018.
- Borsdorff, T., aan de Brugh, J., Pandey, S., Hasekamp, O., Aben, I., Houweling, S., and Landgraf, J.: Carbon monoxide air pollution on sub-city scales and along arterial roads detected by the Tropospheric Monitoring Instrument, *Atmospheric Chemistry and Physics*, 19, 3579–3588, <https://doi.org/10.5194/acp-19-3579-2019>, URL <https://www.atmos-chem-phys.net/19/3579/2019/>, 2019.
- Dekker, I. N., Houweling, S., Aben, I., Röckmann, T., Krol, M., Martínez-Alonso, S., Deeter, M. N., and Worden, H. M.: Quantification of CO emissions from the city of Madrid using MOPITT satellite retrievals and WRF simulations, *Atmospheric Chemistry and Physics*, 17, 14675–14694, <https://doi.org/10.5194/acp-17-14675-2017>, URL <http://dx.doi.org/10.5194/acp-17-14675-2017>, 2017.
- Dekker, I. N., Houweling, S., Pandey, S., Krol, M., Röckmann, T., Borsdorff, T., Landgraf, J., and Aben, I.: The origin of CO sources during the 2017 high pollution episode in India determined with TROPOMI and WRF data, manuscript in prep., 2018.
- Mejia, J. F.: Running WRF in an Atmospheric Modeling Class: challenges and learning experiences, *Atmosfera*, 2020.