

## Review of "Quantifying variability, source, and transport of CO over the Himalayas and Tibetan Plateau" by Sun et al.

This paper presents an analysis of a suite of insitu CO measurements located in the Himalayas and Tibetan Plateau, providing source attributions of the CO measurements through the use of a GEOS-Chem full-chemistry simulation, a GEOS-Chem tagged CO simulation and the HYSPLIT trajectory model. The authors ultimately conclude that main source of CO in the studied region is due to local and Southeast Asian anthropogenic and biomass burning, and oxidation sources. In contrast, black carbon in the studied region is largely attributed to biomass burning of Southeast Asian. They also conclude that there is a decreasing trend in CO due to decreasing emissions in the region.

I believe that the results of this study could be of interest to the general atmospheric science community; however, in its current state, I feel the manuscript lacks sufficiently novel results while there is no new contribution to methods for this type of analysis. The methods are nearly identical to those in the recent paper by Sun et al. 2020. My greatest concern of this paper is on the use of the GEOS-Chem model. The bulk of the main conclusions of the manuscript are derived from interpretation of the GEOS-Chem simulation. However, I believe there are a number of major flaws that must be addressed as I suspect they could likely change the results. I have a number of major comments that should be sufficiently addressed before the manuscript is considered for publication. If the major comments can be sufficiently addressed, I would recommend the paper for review subject to addressing minor issues.

**Major Comments** 1. Perhaps I am missing something but I do not see how CO measurements alone can provide evidence for the authors main motivation that the HTP region is an important region for pollution. As the authors state themselves, the HTP CO is largely attributed to background concentrations due to long-range transport. The results of the analysis also seem to come to this conclusion. The authors had identified cases of enhanced CO measurements which were attributed to biomass burning or local anthropogenic sources. However, I do not think a robust conclusion can be made here. CO has a moderate atmospheric lifetime and therefore can undergo long-range transport. Other atmospheric constituents, that may be more important to pollution in the HTP may have much shorter lifetimes. Therefore, these pollutants may have little to no influence over the HTP. This can not be determined from the CO measurements alone, but the GEOS-Chem full-chemistry simulation could provide some insight. However, this is subject to certain model limitations discussed below.

2. GEOS-Chem is a powerful tool for source attribution studies of this kind; however, I feel that the way the authors implemented the model raises several questions.

In Section 2.2, the GEOS-Chem model setup is described. On L37-38, the authors state a 1hr timestep for surface variables and PBL heights. I am not sure what surface variables are in this case or is the PBL timestep? I am guessing these are the emissions and PBL mixing timesteps? Additionally, given the importance of the PBL in this studies, the authors should state which PBL mixing scheme was used. The authors also state a 3hr timestep of all other variables. Is this referring to transport and chemistry timesteps? If so, this seems exceedingly long, especially for the full-chemistry simulation. The authors should refer to Philip et al. 2017 for a discussion of the appropriate timesteps to be used.

For the tagged CO simulation, the OH fields, methane and VOC oxidation rate are derived from a full chemistry simulation. Is this from the full-chemistry simulation used in this study? I believe this would be the best approach to maintain consistency between the two simulations. If they are not from this simulation, the model version should be stated. On a related note, the authors state the GEOS-Chem version, but it is now required that the model DOI is also include. The authors should add this the manuscript.

Philip, S., Martin, R. V., & Keller, C. A. (2016). Sensitivity of chemistry-transport model simulations to the duration of chemical and transport operators: a case study with GEOS-Chem v10-01. *Geoscientific Model Development*, 9(5), 1683–1695. <https://doi.org/10.5194/gmd-9-1683-2016>

3. In Section 4 "Model evaluation over the HTP", I have concerns about the conclusions that could be made from this type of model evaluation. Particularly, I am unsure why the authors have compared the model to measurements by first scaling the model concentrations based on a mean model to measurement ratio. This effectively eliminates any bias in the model due to OH biases, weak vertical mixing and excessive stratosphere-troposphere exchange, all of which are known issues in the GEOS-Chem model. As a result, I believe these issues could therefore influence the interpretation of the results. Particularly, I would assume that GEOS-Chem would likely capture mostly background concentrations of CO due to long range transport, while episodic enhancements would not be accurately captured, I believe the authors come to this conclusion at the end of the section.

Furthermore, can robust conclusions be made for these comparisons. I believe that high-altitude insitu measurements and valuable, but for model evaluation, it is difficult to assess. Particularly, due to the representativeness error of the insitu measurements only represent a single model level. I would guess that column measurements could provide a more appropriate metric. I understand that column measurements are likely scarce in this region; however, it could be worth considering a comparison to satellite-based CO measurements from instruments such as MOPITT or IASI in a larger domain. 4. In Section 5.3, I believe this method is reasonable, but would the results be more concise if the authors had implemented a transport model such as STILT or FLEXPART? These models could easily identify the source-receptor relationship for the enhanced measurements. Particularly these models can be easily and efficiently run at high resolution, providing better representation of vertical transport than GEOS-Chem.

5. The authors have stated they had used a full-chemistry GEOS-Chem simulation. Aside from the brief discussion of the CO/NO<sub>2</sub> ratios, they results of this simulation are not discussed further and do not contribute to any substantial conclusions. I would suggest to just remove this simulation from the analysis completely, as the authors are only using CO measurements. Alternatively, the authors could include the results of the full-chemistry simulation for other species as I mentioned in the first comment, but this analysis would have to be carefully considered given the model limitations. It may also be outside the scope of the study.

#### **Minor Comments:**

In Section 4: On L30-32, I do not believe the model underestimation is due just to underestimation in local anthropogenic sources. It is likely a contributing factor, but OH biases, weak vertical mixing and excessive stratosphere-troposphere exchange are likely larger contributors.

L38-39, it is unclear what the 0.25 x 0.3125 degree model grid cell is referring to. On L40-42, I am also not sure what is meant by mean concentration.

In Section 5.1 and 5.2, the authors should carefully consider the potential uncertainties in the GEOS-Chem model and discuss the potential impacts on their interpretation of the results.

In Section 5.2 on L26, can the authors explain this seasonality?

FIRMS fire hotspots are used, but would it not be easier and more consistent to just use the GEOS-Chem biomass burning emissions? Fire locations do not necessarily correspond to emissions. On the following lines, what is meant by intensive fire numbers? And again on L2-3 of page 11, why not just use the GEOS-Chem emissions to verify that it is an anthropogenic source?

In Section 5.4, first paragraph, what is meant by self-clean? Could the authors clarify.

In Section 5.4, page 12, L11, I would think OH seasonality is also a major factor.

In Section 5.4, page 12, L14, "accumulation, diffusion and depletion processes", not sure what this means. Please clarify.

In Section 5.4, page 12, L38-40, where does the CO/CO<sub>2</sub> ratio come from? A reference is needed, or this sentence should just be removed.

Figures 2-10, can the authors please ensure that the y-axis of each subplot is consistent? It's very difficult to interpret since all plots have different y-axis ranges.

Figure 11-12, I would recommend to include a colorbar for the CO concentrations.