

Response to Referee #1:

Thanks very much for your comments, suggestions and recommendation with respect to improve this paper. The response to all your comments are listed below. There was an extensive discussion among the authors regarding how to revise the content, and this paper is subjected to a major revision for addressing the concerns by all the referees and Dr. S. K. Sharma. Thus, the response is delayed, and we are sorry for this.

This paper presents an analysis of a suite of in situ 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.

Response: All major comments listed below have been addressed. Please check the point by point response as follows.

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.

Response: The motivation of this study is to quantify variability, source, and transport of CO in the urban areas over the Himalayas and Tibetan Plateau (HTP) rather than to prove that the HTP region is an important region for pollution. Most previous studies have often concentrated on burdens, sources and transport of carbonaceous aerosols (including organic carbon (OC) and black carbon (BC)) over the HTP, but the studies on gaseous pollutants are limited (Cong et al., 2007; Cong et al., 2009; Cong et al., 2013; He et al., 2014; Zhang et al., 2015; Zhu et al., 2019; Li et al., 2021; Gul et al., 2021; Thind et al., 2021). An investigation of CO pollution can complement current atmospheric investigation over the HTP since the chemical characteristic, climate forcing, and deposition of CO is different from the well-established carbonaceous aerosols.

Indeed, we compared our study with previous studies, but this is intended to highlight the novelty of this study. This study only cares about CO pollution in urban areas over HTP.

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.

Response: We apologize for the confusion. The original text in L37-38 are for time step of the input meteorological fields from GEOS-FP, i.e. 1-hour for surface meteorological variables (e.g. surface temperature) and PBL heights and 3-hour for other meteorological variables. This is not the time step for chemistry, emission, and transport. We now state in the Section 2.4 to avoid confusion:

“The time step used in the model are 10 minutes for transport and 20 minutes for chemistry and emissions, as recommend for the GEOS-Chem full-chemistry simulation at 2x2.5 (Philip et al., 2016).”

The time step of PBL mixing follows that of transport (i.e. 10 minutes). We add the following text for the PBL scheme description.

“The non-local scheme for the boundary layer mixing process are described in Lin and McElroy (2010).”

[1] Lin J T, Mcelroy M B . Impacts of boundary layer mixing on pollutant vertical profiles in the lower troposphere: Implications to satellite remote sensing[J]. Atmospheric Environment, 2010, 44(14):1726-1739.

For the tagged CO simulation, the OH fields, methane and VOC oxidation rates 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.

Response: Yes, the OH fields, methane and VOC oxidation rates are derived from a full chemistry simulation used in this study. To maintain consistency between different simulations, the same version of GEOS-Chem, emission inventories and meteorological fields are used. The model DOI is included in the revised version.

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>

Response: We have included this reference in the revised version.

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 in situ measurements and valuable, but for model evaluation, it is difficult to assess. Particularly, due to the representativeness error of the in situ 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.

Response: We have followed your suggestion and used IASI CO total column from 2015 to 2020 over the HTP to evaluate the model performance in the specifics of the HTP. Please see section 4 for details. To balance the accuracy and the number of valid data over HTP, the IASI data within $\pm 1^\circ$ latitude/longitude rectangular area around each city and with total error of less than 15% are selected. Since the IASI overpass time is at about 09:30 LT in the morning, only the GEOS-Chem simulations at 9:00 and 10:00 LT are considered. The results show that, though not perfect in reproducing the absolute values of the IASI observation, GEOS-Chem can capture the measured seasonal cycle of CO total column over the HTP with a correlation coefficient (r) of 0.64 to 0.82 depending on regions. The agreement is much better than using the in situ data.

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.

Response: In order to determine the 3D transport pathways, the 3D travel trajectories, latitude/longitude – height distributions of CO VMR along with the 3D atmospheric circulation patterns have to be used. For example, “CO originating in distant regions such as western EUBA and NA reaches a higher altitude (to 8 km) than those in SEAS region”, “CO removals over the HTP in all seasons are driven by atmospheric deep convection which lofts CO into higher altitudes or by westerlies which transports local emissions far away”, etc., these information cannot be derived without 3D travel trajectories and 3D atmospheric circulation patterns.

Indeed, STILT, FLEXPART and HYSPLIT are all powerful tools to identify the source-receptor relationship for the enhanced measurements, and we used the HYSPLIT to generate 3D trajectories in this study. We believe STILT and FLEXPART can do the same thing but we don't think we can simplify the analysis by using either of them to obtain the same results. Otherwise, we can only get the regional sensitivity (this can be easily done by flatten the trajectories and count the frequencies within each region) but cannot get the 3D transport processes.

5. The authors have stated they had used a full-chemistry GEOS-Chem simulation. Aside from the brief discussion of the CO/NO₂ ratios, the 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.

Response: We apologize for the confusion. The CO and NO₂ time series used here are all based on in situ measurements rather than model simulations. The results show that NO₂ and CO concentrations were correlated in all cities (r ranges from 0.49 – 0.86) throughout the year. The overall good correlations between these two gas pollutants suggested common sources of ΔNO_2 and ΔCO in these cities. As a short lifetime species (a few hours), the emitted NO₂ is heavily weighted toward the direct vicinity of local emission regions. As a result, local emissions are important sources of CO in all cities. We have stated that both CO and NO₂ time series are in situ measurements in the revised version. This section is used to prove that local emissions are important sources of CO in all cities. We would like to keep it in the revised version.

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.

Response: These GEOS-Chem vs. IASI differences over the HTP were mainly attributed to the underestimation of local emission inventories and the coarse spatial resolution of the GEOS-Chem model grid cells. In addition, the difference between simulation and measurement could be also associated with the uncertainties in meteorological fields over HTP, OH field, and stratosphere-troposphere exchange (STE) scheme, which are known issues in the GEOS-Chem model (Bey et al., 2001; Kopacz et al., 2011). Please see detailed explanation in section 4.

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

Response: It should be 2×2.5 degree model grid cell and we have reworded this sentence to avoid misleading, i.e., “Besides, the coarse spatial resolution of the GEOS-Chem simulations homogenizes CO concentrations within each $2^\circ \times 2.5^\circ$ model grid cell. The simulation results represent the homogenized concentrations in the grid box at the grid-mean elevation, which could cause significantly bias near complex terrain (Yan et al., 2014)”.

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.

Response: We have discussed the potential impacts on the interpretation of the results

in the revised version. Please check section 4 and section 5.2 for details. “ We turn off all emission inventories within the HTP in the GEOS-Chem tagged CO simulation and assess the relative contribution of each source and geographical tracer. The relative contribution of each tracer is calculated as the ratio of the corresponding absolute contribution to the modelled total amount. Taking this ratio effectively minimizes the propagation of systematic model errors that are common to all tracers, i.e., the uncertainties in meteorological fields, the vertical mixing and STE schemes, and the mismatch in spatial resolution.”

“By minimizing the propagation of model errors that are common to all tracers (see section 4), the major factors impacting the model interpretation are the uncertainties in emission inventories and OH fields. The uncertainties in CO emission inventories mainly impact primary anthropogenic and BB sources, and the uncertainties in CH₄ and VOCs emission inventories, and OH fields mainly impact secondary oxidation sources. Additional factors that affect the generation and depletion chemistry of CO or its precursors (e.g., uncertainties in emission inventories of other atmospheric components, stratospheric intrusion of ozone and chemical mechanism, etc.) could also contribute to the uncertainty of the interpretation. All these factors may be seasonal and regional dependent. A series of GEOS-Chem sensitivity studies might be able to quantify these uncertainties, but this is beyond the scope of present work.”

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

Response: We have explained the seasonality in the revised version, i.e., “The JJA/SOJ meteorological conditions which show stronger solar radiation, higher temperature, wetter atmospheric condition, and lower pressure than those in DJF/MAM are more favorable for increasing VOCs emissions from biogenic sources (BVOCs), which consolidates the fact that contributions of NMVOCs oxidation in warm season are larger than those in cold season.”. Please check section 5.2 for details.

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?

Response: We have followed your comments and used GEOS-Chem emissions to identify anthropogenic and biomass burning sources.

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

Response: We have defined the terminology of self-clean, i.e., “Atmospheric

self-clean capability refers to the capability of the atmosphere in terms of depleting atmospheric pollutants through physical and chemical processes (Rohrer et al., 2014)". Please check section 5.4 for details.

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

Response: We have changed it to "The OH oxidation capability is positively correlated with temperature, radiation and OH seasonality". Please check section 5.4 for details.

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

Response: We have changed it to "generation and deplete processes".

In Section 5.4, page 12, L38-40, where does the CO/CO₂ ratio come from? A reference is needed, or this sentence should just be removed.

Response: We have removed this sentence in the revised version.

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.

Response: We have used consistent y-axis for most of these figures except the sub-figures that we intend to clearly show seasonal variation of each quantity.

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

Response: Done.