Response to Referee #2:

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.

The authors analyzed the site measurements and modeling results of CO over the HTP in terms of its diurnal and seasonal cycle as well as interannual variability. They also used GEOS-Chem and HYSPLIT to identify the major long-range transport path of CO globally to the HTP. The analysis was mainly done for several urban regions in the HTP. In general, the manuscript structure is well laid out, but there are still a few places requiring further clarification and improvement. My suggestions and comments are as follows.

Major comments:

1. My major concern is the relatively low model spatial resolution for GEOS-Chem (2deg) and HYSPLIT (1deg) simulations, which could not capture the complex topography over the HTP. Particularly, this might miss some efficient valley-mountain transport. How would the spatial resolution issue affect the transport and variation analysis in this study? Besides, it is also a problem when evaluating GEOS-Chem model by comparing 2deg grid value with point-scale site measurements.

Response: Indeed, GEOS-Chem (2deg) and HYSPLIT (1deg) are probably not good enough for local emission, but it is sufficient for investigation of long range transport in this study.

As recommended by referee #1, we have used IASI CO total column from 2015 to 2020 over the HTP to evaluate the model performance in the specifics of the HTP. To balance the accuracy and the number of valid data over HTP, the IASI data within ±1° latitude/longitude rectangular area around each city and with total error of less than 15% are selected. Indeed, the extreme climate conditions (e.g., valley-mountain transport) and the variability between clean and polluted conditions in the region are a challenge for current climate models. The comparison shows 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. GEOS-Chem can capture mostly background concentrations of CO due to long range transport but cannot accurately capture episodic enhancements. In this study, the GEOS-Chem model is only used for investigating the influence of long range transport. 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 minimises 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. Please see section 4 for details.

2. Section 5.3: It's interesting that the authors combined GEOS-Chem simulation and HYSPLIT back trajectory analysis to identify the path. But note that the meteorological fields used in GEOS-Chem and HYSPLIT are different, which could lead to some inconsistency. Maybe a brief comparison of wind fields for these two would be useful.

Response: In the revised version, we verified that the wind fields provided by GDAS-1 are in good agreement with those by the Goddard Earth Observing System-Forward Processing (GEOS-FP) meteorological fields used in GEOS-Chem (Fig. S1). Please see supplement for details.

3. One important thing that was not discussed by the authors is how the uncertainty in VOC emissions contributes to the uncertainty in the analysis here, given the non-trivial contribution from secondary CO production. Besides, how would the stratospheric intrusion of ozone which is important over the HTP affect the CO simulations here?

Response: This is a very interesting topic but can't be solved through a simple analysis. A series of GEOS-Chem sensitivity studies might be able to quantify these uncertainties, but this is beyond the scope of present work. Please see the following discussion in section 5.2 in the revised version.

"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 CH_4 and VOCs emission inventories, and OH fields mainly impact secondary oxidation sources. Additional factors that affect the generation and deplete 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."

Minor comments:

1. It seems that this study mainly focuses on urban areas over the HTP, so I suggest changing the title to reflect this aspect to avoid confusion, since conclusions here may not be applicable to remote areas in the HTP.

Response: We have changed the title to "Quantifying variability, source, and transport of CO in the urban areas over the Himalayas and Tibetan Plateau".

2. Page 3, Line 8: Note that BC is also one aerosol component, so the authors could be more specific about the aerosol here. Also, a few important recent studies can be included here, for example, Li et al. (2021): https://doi.org/10.1016/j.envint.2020.106281; Gul et al. (2021): https://doi.org/10.1016/j.envpol.2021.116544

Response: These important recent studies have been included in the introduction. "These atmospheric compositions include CO (Park et al., 2007a; Park et al., 2007b), CH4 (Xiong et al., 2009), HCN (Randel et al., 2010), PAN (Zhang et al., 2009; Ungermann et al., 2016; Xu et al., 2018), O_3 (Yin et al., 2017; Xu et al., 2018), and aerosol (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)."

Since some of these studies described more specific about the aerosol but some are not. As a result, we grouped all of them as aerosol as above. Please see section 1 for details.

3. Page 3, Lines 21-22: ". . . are still poorly understood". I think the community has made important advances in the past 10 years on this topic over the HTP. Many studies have investigated the sources and transport of atmospheric pollutants over the HTP, although many of them have used black carbon instead of CO as a tracer (e.g., Thind et al., 2021: https://doi.org/10.1016/j.atmosenv.2020.118173; He et al., 2014: https://doi.org/10.1002/2014GL062191; Zhang et al., 2015: https://doi.org/10.5194/acp-15-6205-2015; Zhu et al., 2019: https://doi.org/10.5194/acp-19-14637-2019). I think these studies have also helped to improve our understanding of pollution transport in this region and could be mentioned in the introduction and compared with the transport results based on CO tracers in this study in the discussion section.

Response: We have revised the statement "…are still poorly understood" and described the recent studies shown above in the introduction, i.e., "… In addition, 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). As a result, the variabilities, sources, drivers, and transport pathways of atmospheric pollutants over the HTP are still not fully understood". Please see section 1 for details.

We also compared our study with previous studies, i.e., " From section 5.1 and the model interpretation here, we conclude that main source of CO in urban areas over HTP is due to local and SEAS anthropogenic and biomass burning emissions, and oxidation sources. In contrast, black carbon in most of the HTP is largely attributed to Southeast Asian biomass burning, and locally sourced carbonaceous matter from fossil fuel and biomass combustion also substantially contribute to pollutants in urban cities and some remote regions, respectively (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). Our study emphasized the different origins of diverse atmospheric pollutants in the HTP.". Please see section 5.2 for details.

4. Page 3, Lines 38-44: One thing that was not mentioned by the authors is that what is the new aspect of this study to look at CO over the HTP compared with previous studies. It seems that not much has been described in the introduction section regarding what knowledge of CO transport in this region we already obtained from previous studies. Some descriptions are needed and the novelty of this study needs to be highlighted.

Response: We have highlighted new aspect of this study to look at CO over the HTP compared with previous studies in the introduction, i.e., "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). CO study 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."

We have also described in the introduction regarding what knowledge of CO transport in this region we already obtained from previous studies, i..e, "Only few studies have investigated the burden and variability of CO over the HTP (Ran et al., 2014; Yin et al., 2019a). These studies uniformly focused on the most developed regions in Lhasa, and did not analyze interannual trend and transport of CO. This study can not only expand the coverage of CO study over the HTP, but also provides insights into the interannual trends, sources, and transport of CO in all urban areas over the HTP.". Please see section 1 for details.

5. Page 12, Line 4: "Factors drive . . ." should be "Factors driving".

Response: Done.