

## **Reply to Referee #2:**

We would like to thank anonymous referee #2 for detailed comments and suggestions that helped us to improve the manuscript. We have carefully considered these comments in our revision of the manuscript. Our responses are provided below (the reviewer's comments are shown inline in italics).

### *Major comments:*

*1. The authors proposed to categorize the upward transport of CO from the surface to the tropical UT into three different categories: local convection, LT advection, and UT advection. If the ultimate goal of this study is to understand how seasonality in surface emissions and dynamic transport contribute to the seasonality of CO in the tropical UT, isn't the three categories too much a simplification in a region where south-north movement of ITCZ, east-west transport associated with ENSO and long-range transport from Indonesia and South Asia, etc, all mingled together? At minimum, the authors need to provide a comprehensive review of what are the dominant processes that could contribute to the observed seasonality and how to interpret their derived results in this larger context.*

**Reply:** This paper aims to identify CO transport pathways from the surface to the upper troposphere (UT), either inside or outside of the CO source regions by convection. The former is represented by the “local convection” transport pathway, whereas the latter is represented by the “lower-troposphere advection and then convection” pathway. These two transport pathways broadly represent long-range transport of CO, including those influenced by ENSO and the ITCZ. Following recommendation made by this comment, we have added a comprehensive review of previous work and distinction between our work and those previous studies in the introduction of the revised manuscript as follows:

“Previous studies based on observations and/or modeling (Novelli et al., 1998; Novelli et al., 2003; Edwards et al., 2006; Duncan et al., 2007a; Liu et al., 2010; Macdonald et al., 2011) have shown that CO seasonal and interannual variability in the troposphere is affected by many factors, which can primarily be divided into the categories of photochemistry and transport. These factors vary greatly among different geographic regions. Edwards et al. (2006) used five years of CO data from MOPITT to show that the interannual variation of tropospheric CO in the southern hemisphere correlates well with the El Niño–Southern Oscillation precipitation index. Duncan et al. (2007a) evaluated the global budget of CO from 1988 to 1997 using a chemical transport model and found that emissions were largely constant during this period, as increases in Asia were offset by decreases elsewhere. Liu et al. (2010) used model simulations to interpret the spatial and temporal variations of CO in the LT and UT, and suggested that prevailing subsidence during the peak fire season over South America (July–August) may trap CO near the surface until convection associated with the inter-tropical convergence zone (ITCZ) moves into the fire region

during September and October. Macdonald et al. (2011) investigated the relative influences of trans-Pacific transport and emissions from North American forest fires on CO concentration at a high elevation site using 5 years of in-situ data. They argued that although trans-Pacific transport plays a role during boreal winter through spring, biomass burning is the most important contributor to periods of elevated CO concentration during summer.”

The contribution of our study is to propose a more systematic approach for identifying CO transport pathways, and to provide a statistical description of the geographic and seasonal variations in these transport pathways. The results enhance understanding of (1) how fire-generated CO is transported from the surface to the UT and (2) the relationships between seasonal changes in pathway prevalence and CO in the tropical UT.

*2. By averaging emission data and satellite CO data at 8-day intervals and  $4^{\circ} \times 8^{\circ}$  (approximately 450 km x 900 km) and using the co-occurrence of surface emission and elevated UT CO as identification of local convection, the authors are in fact assuming air remain relatively stagnant in the region. This in fact might not well be true. Assuming mean winds about 5 m/s in the LT and 20 m/s in the UT (which are reasonable numbers), an air mass can travel ~3500 km and ~14000 km in 8 days and be placed in a distant downwind region. Therefore, while surface emissions indicate surface fire activity and satellite data shows elevated CO above in the UT, it is some times possible that the elevated UT CO is not associated with local convective lofting, but advection from remote resources.*

**Reply:** This point is well taken. In our view, using 8-day average does not imply that air remains relatively stagnant in each region, although we agree that this approach would underestimate CO increases associated with convective transport because the CO change associated with advection is much smaller than that associated with convection, as shown by Fig. 9 in our revised manuscript. Nevertheless, we have replaced our previous approach with one based on the transport pathways identified using instantaneous along-track co-located measurements of Aura MLS and CloudSat. The main conclusion of the original manuscript, i.e., the strong correspondence between the seasonality of CO concentrations in the tropical UT and the seasonality of the “local convection” transport pathway, is still valid based on this new analysis. Furthermore, the seasonal pattern of “local convection” is effectively unchanged when MERRA reanalysis data is used to eliminate cases with UT convergence (the presence of which might indicate advection from remote sources in the UT rather than local convective lofting).

*3. Although the authors did not say explicitly, the discussion in the second paragraph on page 32426 and section 5 seems to imply that emission and transport are the only processes that contribute to seasonality*

*of CO. This is misleadingly incomplete. A significant part of CO seasonal variations is also due to i) seasonal changes in its lifetime due to changes in solar radiation (therefore OH) (Duncan et al., 2007), ii) seasonal changes in CO production from biogenic sources and CH<sub>4</sub> oxidation (Duncan et al., 2007; Liu et al., 2010). The impact of the above, relative to seasonal variations due to convection transport and surface emissions, need to be addressed.*

**Reply:** This point is well taken. We have added discussion on the factors contributing to CO seasonality. In addition to the discussion described in our reply to major comment 1, we have also added the following discussion in line 427–447 on page 20-21 of the revised manuscript:

“The lifetime of CO in the troposphere is largely determined by the concentration of OH, its main sink (Levy, 1971; Thompson, 1992). The levels of tropospheric hydroxyl radicals are strongly correlated with solar ultraviolet radiation (Rohrer and Berresheim, 2006) and have different seasonal cycles at various latitudes. The lifetime of CO in the troposphere is longer during local winter than during local summer, especially in the middle and higher latitudes (Duncan et al., 2007a). Because this work focuses on the tropical (20°S–20°N) UT (215 hPa), seasonal changes in CO lifetime are therefore much less than those in the mid-latitudes.

Biomass burning represents the largest source of CO in the tropics during burning seasons, especially in the SH; during other seasons, oxidation of CH<sub>4</sub> and biogenic non-methane hydrocarbons (NMHC) may be more important (e.g., Logan et al., 1981; Holloway et al., 2000; Duncan et al., 2007a; Liu et al., 2010). The seasonal variations of CO production from biogenic sources and CH<sub>4</sub> oxidation at the surface and in the LT are also relatively small compared to the seasonal variations of biomass burning in the SH (Duncan et al., 2007a; Liu et al., 2010). In the UT, Liu et al. (2010) used model simulations to show that seasonal changes in the biogenic source (from isoprene) do play an important role in the seasonality of UT CO over South America; however, biomass burning is the largest source of CO to the UT in burning regions during the biomass burning seasons (November–February in the NH, June–October in the SH). Therefore, the seasonal variations of CO in the tropical UT are likely to be largely determined by convective transport of CO generated by biomass burning.”

*4. The authors state that “To our knowledge, the influences of seasonality in the distribution of transport pathways on the seasonality of CO concentration in the tropical UT have not yet been clearly identified or addressed.” The two above referenced papers, Duncan et al. (2007) and Liu et al. (2010), though did not address directly the impact of the distribution of transport pathways, both presented a comprehensive modeling analysis together with satellite measurements to look at the impact of upward transport of biomass burning emissions as well as many other sources on the seasonality of CO in the UT and LS region. These two studies are highly relevant to this study and need to be acknowledged. In addition, I*

*strongly encourage the authors to compare the results from this work with those from Duncan et al. (2007) and Liu et al. (2010) and discuss how they agree or disagree.*

*For example, the authors conclude that “This result suggests that the seasonal prevalence of the local convection pathway plays a key role in the seasonal variation of UT CO over Central Africa” and “the seasonal variation of UT CO over South America was consistent with that of the occurrence frequency of the local convection pathway” (Page 32436). In comparison, Liu et al. (2010) suggested that the spring and fall peaks in CO at 215 hPa over tropical Africa are due to the combined impact of local biomass burning emissions, long-range transport from Indonesia and South America, as well as production from biogenic emissions. Similarly, the austral spring peak in South America is due to the combined impact of local biomass burning emissions, long-range transport from S. Africa and production from biogenic emissions (Liu et al., 2007).*

**Reply:** Thanks for this helpful comment. We have added more discussion on these two works in line 110–127 on page 5-6 of our revised manuscript as follows:

“Duncan et al. (2007b) studied troposphere-to-stratosphere (TST) transport of CO and evaluated the impacts of biomass burning pollution on the composition of the tropical tropopause layer (TTL) and lowermost stratosphere (LMS). They found that two TST pathways (slow ascent through the TTL and quasi-horizontal exchange into the LMS) are of similar importance for the transport of biomass burning pollution. By contrast, our study focuses on the pathways that transport CO from the surface to the UT (215 hPa, below TTL) and how their seasonal variations influence the seasonal variation of UT CO over two important biomass burning regions. Liu et al. (2010) used GEOS-Chem model simulations to evaluate CO transport in the GEOS-4 and GEOS-5 assimilated meteorological fields and to identify causes of discrepancies between observations and simulations.

This study develops a method using co-located satellite measurements to automatically identify the two convection-induced CO transport pathways, and uses this method to study the relative importance of these two pathways to seasonal variations of CO in the tropical UT. Although CO concentration can be influenced by multiple transport pathways simultaneously, the two transport pathways defined here broadly represent long-range transport of CO from the surface to the UT, such as that associated with the ITCZ.”

*Minor comments:*

*Section 3.1 – the Boreal winter case: By looking at the wind arrows, it seems the northward transport at ~11km is ~5 times faster than the near-surface southward transport. This implies if the transport pathway is LT advection->convection-> UT northward advection as the authors argued, the CO hot spot in the UT should be displaced to the north of the surface emission center, not where convection happens. What is*

*the possible explanation in the UT displacement? In addition, I suggest the authors include a figure showing back trajectory results for this case. This can be a more robust piece of evidence in demonstrating the transport pathway.*

**Reply:** The above issue is a result of the 8-day averages in our original manuscript, i.e., during which the 8-day mean convection and wind patterns may be contributed by these fields from different days. In our revised manuscript, we use instantaneous along-track data, thus this problem no longer exists.

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