

Reply to Referee #1:

We would like to thank anonymous referee #1 for detailed comments that helped us to improve the manuscript. We have carefully considered each of the reviewer's comments in our revision. Our responses are provided below (the reviewer's comments are shown inline in italics).

*Major comments:*

*1. After reading the manuscript, I am lost of what the main scientific questions are. Yes, even if you do a good job showing the contribution of these three pathways, what usefulness of these three pathways? What are the questions you want to address?*

Reply: As discussed in the introduction section and Fig. 1, the seasonal cycle of CO in the upper troposphere (UT) over tropical (30°S–30°N) land areas differs from the seasonal cycles of both CO emission and convection. This indicates that insight into how fire-generated CO is transported from the surface to the UT is important for understanding the seasonality of CO in the UT. Although previous studies have shown the existence of a variety of possible pathways (e.g., Pickering et al., 1996; Folkins et al., 1997; Andreae et al., 2001; Williams et al., 2002; Andreae et al., 2004; Ray et al., 2004), no systematic evaluation of their relative importance in determining the seasonal cycle has yet been performed. This study provides such an evaluation, and addresses questions regarding how different transport pathways affect the seasonal and geographic distributions of CO concentration in the UT. The automated pathway-identification method that we present in this paper generally captures the occurrence of convective transport of CO to the UT, both within (“local convection”) and outside of (“LT advection → convection”) fire regions. These two pathways are primarily responsible for delivery of CO to the UT. The third pathway considered, the “UT advection” pathway, mainly represents the redistribution of CO within the analysis domains. As such, it is important to our detailed interpretation of the geographic distribution of CO in the UT, but it does not impact our assessment of how CO is transported from the surface to the two centers of high UT CO that we analyze in detail (South America and Central Africa). This method represents an important step toward understanding what controls the seasonal cycle of CO in the tropical UT.

*2. The direct transport pathway by convection is easier to identify, but other two pathways LT-Convection and UT advection are relatively difficult, if not possible, to separate. Using 8-day average could smear many things out. Explanation of the first case by LT-Convection is very difficult to believe especially when you are inferring 1000 km horizontal transport without significant mixing.*

Reply: The difference between “UT advection” and “LT advection → convection” is whether convection is present. Deep convection is accompanied by upper level divergence, which acts to transport CO away

from its convective source. “UT advection” is therefore unlikely to be the primary reason for an increase in UT CO when deep convection occurs. To address the reviewer’s comment, we can evaluate the horizontal gradient of the UT CO to identify and flag those cases when local UT CO is less than that in the surrounding regions as the cases in which “UT advection” could cause an increase of UT CO even in the presence of active deep convection. The 8-day timescale represents a trade-off between (1) improving observational sampling and accuracy and (2) smearing out the effects of different transport pathways. The pathways identified using this timescale are consistent with those identified by a much more intensive trajectory analysis using 6-hourly winds, suggesting that our approach and chosen parameters are reasonable.

*3. Using cloudsat as the presence of deep convection is problematic. Remember CloudSat only provide a thin vertical cross section. More often it only crosses the edges of a large MCS. Therefore, it is difficult to use it as a direct measure of the strength of convection. There are a lot of cirrus clouds contributing CWC that are not necessarily from deep convection. Sampling is another problem. If you plot the samples from CloudSat daily, the holes are huge globally. I am not sure if 8-day cloudsat average can give a good representation of the convection, even in a big 4x8 grid.*

Reply: It is true that CloudSat may not detect the strongest convective events due to its orbit setting, and that it will miss a number of convective transport events. We have performed this analysis twice more, independently using the TRMM cloud feature database (Liu et al., 2008) and the ISCCP D2 deep convective cloud amount dataset (Rossow and Schiffer, 1999) as indicators of convective activity in lieu of CloudSat CWC. The 8-day and seasonal maps are nearly identical in all three cases. Cirrus clouds that are not associated with convection typically have a much smaller CWC than convective anvils captured by our CWC threshold. CloudSat CWC appears to be adequate for determining the large-scale distribution of convective activity on 8-day and seasonal timescales.

*4. The scenario of LT advection-convection is very ambiguous. When there is increasing UT CO, with convection, but without high surface CO, it must be LT-convection? Logically, it can also be quite possible a UT advection case. I would rather believe latter, unless you can provide evidences to rule out the UT advection by using upper level wind and low upstream CO.*

Reply: As described in our reply to Comment 2, this scenario is possible, but unlikely. We have added a criterion that requires the local UT CO to be higher than that in the surrounding regions.

*5. The method of analysis is not quantitatively described. What determine UT CO increase? What CWC determine convection? What determine surface CO? These are very important to understand the results.*

Reply: This is a very helpful comment. We have added a fuller, more quantitative description in the revised manuscript: We use the difference of CO concentrations at 215 hPa between two consecutive 8-day periods to determine UT CO increase (this is also mentioned in the manuscript). The threshold of UT CO increase used in the manuscript is 1ppbv. The results are qualitatively robust to a ranged of thresholds from 1–10 ppbv. Changing the threshold only affects the number and percentage occurrence of transport events and pathways; it does not affect the relative importance of the transport pathways and their seasonal pattern. Considering the MLS CO measurement uncertainty, we have changed this threshold to 18 ppbv in our revision, and the seasonal pattern remains the same as that in our original manuscript. We used probability density functions (PDFs) to determine the thresholds for deep convection (CloudSat CWC) and surface CO emission (MODIS fire counts). The threshold of CWC is  $100 \text{ mg m}^{-3}$ , which is the median value of all the CWC over the tropical South American and Central Africa domains. This CWC threshold is substantially higher than that expected for in situ cirrus clouds. The threshold of fire counts is 10  $1 \text{ km} \times 1 \text{ km}$  pixels within the grid cell. This threshold is equal to the median value of fire counts over tropical land areas.

*6. The proof of the success of the classification is weak (see minor comment #10). Though the final results seem somewhat reasonable in certain ways, better validation of the method is still needed.*

Reply: The success of the method is quantified by the fourth column (“TrjFrac”) of Table 1 and the discussion in the text. Agreement of 40%+ in every case suggests that even when the method does not identify the predominant (majority) pathway, it still identifies a prevalent pathway. Upon reflection, the third column of the table (“Trajectory pathway”) may be misleading to the reader who doesn’t have time to examine the text in detail. We will remove the third column of the table in our revision, so that it only says what fraction of trajectories agrees with the method’s designation. Readers can then make their own judgment as to whether the automated detection and the trajectory detection agree sufficiently.

*7. Though the focus is on South America and Africa, it is still useful to show how the classification works globally. It is another way to test if the method works or not.*

Reply: Our work focuses on South America and Africa because they are the main centers of high tropical UT CO and dominate the seasonal cycle of tropical mean UT CO. The method itself is not region specific, and should be applicable to other regions. As described in the text, CO emission in some regions outside of these two domains is contributed substantially by fossil fuel burning, so that CO emission data based on MODIS fire data may not adequately represent the distribution of CO emission at the surface. We focus on land areas because CO emission and the centers of high UT CO in the tropics are both generally located over land areas.

*Minor comments:*

*1. 32424, line 8, be specific on these three pathways. It is kind of confusing in the current abstract.*

Reply: Thank you for pointing this out. We will clarify this by revising the text to read: “two pathways that transport CO from the surface to the UT (“local convection” and “advection within the lower troposphere (LT) followed by convection”).”

*2. 32424, line 13, define LT.*

Reply: This is addressed by our revision in response to minor comment 1.

*3. 32424, line 17, highest CO near surface or at upper troposphere?*

Reply: Revised to: “The centers of high CO in the UT ...”.

*4. 32425, line 27, citations needed for the case studies*

Reply: Revised to: “...previous case studies (e.g., Pickering et al., 1996; Folkins et al., 1997; Andreae et al., 2001; Williams et al., 2002; Andreae et al., 2004; Ray et al., 2004) ...”.

*5. 32429, line 5-7, I do not think averaging 8 days NCEP data is a good idea. 8 days is enough for a major transition of atmospheric environment. The mean wind field could be very misleading.*

Reply: The mean wind fields should be representative of the prevailing conditions, and are only used for illustrative (rather than diagnostic) purposes.

*6. 32429, How much confidence do you have on the CWC above 6 km representing the deep convection transporting CO to 215 hPa and 100 hPa? There are many cirrus clouds can give you CWC, but have nothing to do with the deep convection.*

Reply: See our reply to major comment 3.

*7. 32429, what are the criteria that you use to define a) increase of CO (by increase 10% from the mean CO for example, or just greater than the earlier 8 day period?) b) deep convection, what CWC value represents deep convection? c) surface CO emission, what value of surface CO is defined as presence of CO emission?*

Reply: See our reply to major comment 5.

8. 32430, line 12-25, what are the strength of low level winds during the event? Why makes you think CO was advected southward? What strength of southerly flow can transport CO 1500 km within 8 days? Also how much horizontal mixing could that be during the horizontal transport at low levels? The local convection can very well transport the local CO emission to upper troposphere make a bull's eye near Angola. Keep in mind that you do not need a strong surface emission to give high values of CO in UT given a healthy deep convection.

Reply: A wind speed of  $2.2 \text{ m s}^{-1}$  would transport CO >1500 kilometers in 8 days. While it is possible that a weak local CO emission could contribute to the observed increase of UT CO, the TES observations of CO in the lower and middle troposphere indicate that CO advection due to strong cross-equatorial winds from the major fire region north of the equator contributed substantially to elevated CO concentrations below the major UT CO center.

9. 32431, line 17, what convective source do your mean?

Reply: Revised to "...identified according to the instantaneous MERRA 3-D convective cloud fraction." Because of a gap in the ISCCP DX data over our case study region and period, we use MERRA convective cloud fraction here because of its consistency with the winds and diabatic heating fields used to calculate the trajectories.

10. 32431, Table 1 shows me that when there is high CO around at 215 hPa between 0-20S, your classification is only correct 3 out of 5. I am not sure if this is successful.

Reply: Please refer to our reply to major comment 6. We will remove the third column of the table in our revision, so that the emphasis is on the fraction of trajectories that agree with the automated method's designation, rather than on a binary identification of the predominant (majority) pathway.

11. 32433, How do you determine dominant pathway? 40% of cases as one class?

Reply: The dominant pathway is determined as the pathway that occurs most often of the three. We will clarify this in our revision.

12. 32434, line 22-25. Southeast Asia is a good place to test if your classification works or not. I am interested to see what kind of results you get. UT advection should be dominant over a large area due to the strong anti-cyclone above.

Reply: Please refer to our reply to major comment 7. This analysis will not be possible until a reliable CO emission dataset that includes biofuel and fossil fuel emissions is made available.

13. 32436, fig 9, how about UT advection? No change of CO by UT?

Reply: As we have discussed in the original manuscript, the “UT advection” pathway represents the redistribution of CO within the UT rather than the introduction of new CO, so it is not considered in this figure. We will clarify this by discussing only the transport pathways that transport CO from the surface to the UT, namely “local convection” and “LT advection → convection”.

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