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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Model study of the cross-tropopause transport of biomass burning pollution" *by* B. N. Duncan et al.

B. N. Duncan et al.

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Response to Anonymous Referee #1

We are grateful for the careful comments of the reviewer. To strengthen our argument and address several of the reviewer's concerns, we have conducted two additional simulations, in which we separate the component of the seasonal oscillation of CO in the UT/LS due to variations in transport (i.e., convection and slow ascent from the TTL to the LS) from the variations in the individual contributions of each CO source. The first is a tagged CO simulation, which gives the breakdown of the total CO from various sources (e.g., methane oxidation, fossil fuels in E. Asia, biomass burning in S. America, etc.) in the UT/LS. The second is a uniform tracer simulation, in which we emit CO homogeneously over the earth's surface (2400 Tg CO/yr) and assume a uniform 25 day lifetime. Consequently, the uniform tracer is decoupled from the seasonal variation in sources, leaving only the variation due to transport. The results of these two simulations clearly show that both variations in CO sources and dynamics are important players in the composition and seasonal variation of CO in the tropical UT/LS.

In addition, we now present a comparison of CO from our simulation with MLS data in the UT/LS. We had access to only a few days of released data of the MLS (version 2/level 2) data product. Since the submission of this manuscript, we have obtained 100+ days of recently released data from the MLS team. We added a comparison of the MLS observations and simulated CO for March at 68, 100, 146, and 215 mb, which shows that the model does a reasonable job reproducing the horizontal distribution of observations at all levels. We also show a histogram of simulated and observed CO from 20N-20S for four months, which illustrates that our simulation has a similar distribution of CO as the observations. Overall, the comparison is favorable, lending confidence to our simulation. We also added a discussion of the limitations of the observations for the purposes of our study, such as the coarse vertical resolution and patchiness of the observations.

The major modifications to this revised manuscript are in 1) Section 5.2-3, where we discuss sources of variation in the UT/LS composition, 2) Section 3.3, where we include a comparison of MLS observations with our simulated CO, and 3) Conclusions.

$>$Duncan et al. present a model study of the transport pathways of biomass burning pollution - essentially carbon monoxide - to the stratosphere. They use a chemical transport model coupled to a GCM which was forced with sea-surface temperatures of 1994-1998. The paper presents a wealth of information and interesting results.

We appreciate the positive feedback.

$>$However, I believe the structure of the paper, and the layout of the results, could be improved. I recommend publication in ACP subject to the following concerns.

ACPD

7, S1614–S1620, 2007

>From the beginning on, the reader is asserted that deep convective transport may not be overly important for transport across the tropical tropopause. The papers quoted to support that assertion, notably the Fueglistaler et al. [2004] study, however, have *not* demonstrated that deep convective transport is not of importance under any circumstances. Rather, they - and subsequent work by Fueglistaler et al. [2005] - showed that no special mechanisms of dehydration tied to deep convection need to be invoked to understand stratospheric water vapour.

We removed the Fueglistaler et al. [2004] reference from the paragraph in the introduction discussing convection penetrating the tropopause.

>For a tracer like carbon monoxide, however there may be a different story. In fact, carbon monoxide is used frequently to study deep convective outflow. Thus, the present paper should demonstrate, rather than assume a-priori, that the transport as represented by the model parameterization of convection is sufficient to explain observations.

We added the following paragraph to the conclusions in Section 6:

"Liu and Zipser (2005) and Rossow and Pearl (2007) found that very few tropical convective systems penetrate the tropopause, so this pathway is not likely important for CO and other trace gases with lifetimes longer than the time-scale of transport to the tropopause via slow ascent. Convection in our simulation did not penetrate the tropical tropopause, yet our simulation reproduces the distributions of MLS observations reasonably well; that is, this pathway is not needed to explain the large-scale features of the observations. However, it may become important locally near convective systems that do occasionally penetrate the tropical tropopause, such as those over central Africa, Indonesia, and South America (Liu and Zipser, 2005)."

>It appears also somewhat surprising that model results are not directly compared to (MLS) observations in the TTL.

ACPD

7, S1614–S1620, 2007

Interactive Comment

See the above discussion. We cannot create seasonal averages as the reviewer suggests because of the lack of data to create an average for most months. There are ˜10 days of observations to create monthly averages in February, March, and September 2005. In time, more data will be released, allowing us to do a more comprehensive comparison. For guidance on the use of the MLS CO data product, we consulted with Nathaniel Livesey and have added him as a co-author on the manuscript.

>Finally, I consider the structure of the paper somewhat less than optimal.

We believe this comment on the paper structure to be a matter of style, which does not effect the presentation of our message. We start with the Indonesian fires simply because it is the most dramatic burning event ever recorded; our goal is to show to the reader up front that the impact of the pollution in the UT/LS can be quite substantial, something that has not been shown before to our knowledge.

>It is certainly not the authors' fault that currently there is no consensus definition of the 'TTL'. However, I'd suggest to replace the awkward 'TTL/LS' with the more widely used 'UT/LS'.

Done.

>In the abstract, and elsewhere, you postulate that 'convection was stronger' during the El-Nino phase. I assume this does not mean 'more vigorous', i.e. higher, but presumably refers to the strength of the Hadley-cell and can be seen in the streamfunctions? Is this really so? You may also want to check for publications searching for such a signal in analysed data.

We have reworked the discussion on the impact of El Niño on the transport of CO to the UT/LS in Section 5.3 and Figure 19. In short, we found that the zonal mean convective updraft flux at 300 mb in our simulation is about 10% higher during the 97-98 El Niño. However, the flux over the tropical eastern Pacific Ocean is 2-3 times higher. The El Niño causes a shift in the location of deep convection, bringing the

ACPD

7, S1614–S1620, 2007

Interactive Comment

convection closer to the tropical source regions (i.e., biogenics, biomass burning, etc.) in S. America and Africa. In effect, more CO from these sources reaches the UT/LS as they are transported more rapidly to the UT by convection than in other years. Our tagged CO simulation clearly shows that the interannual variability in the location of deep convection plays a large role in the interannual variability of the composition of the UT/LS.

> p2200/l16: You may want to be a bit more specific rather than saying 'etc.'. Also, Haynes and Shuckburgh [2000] may be a useful reference with regards to transport across the subtropical jets.

Thank you for the reference. We included it.

> p2203/l18: Certainly, specifying a seasonal cycle in anthropogenic emissions, even if north of 35 degrees, is a delicate thing to do in the context of explaining the CO tape recorder, and a few words about its impact may be appropriate here.

The seasonal variation of CO emissions from transportation in the extra-tropics is a well known phenomenon, which amplifies the annual cycle in tropospheric CO in the N. Hemisphere associated with the seasonal variation in OH. We now discuss this in the text.

> p2207/l14: I am not sure I understand what you say here - why is the lifetime 'weighted toward the lower tropical troposphere'? I assume that for each level there is a different lifetime?

We clarified this sentence: "Though the lifetime is reasonable in our model, it does not provide information on the quality of model OH outside of the lower tropospheric tropics; the rate constant for reaction of OH with CH3CCl3 is strongly temperature dependent. This is an important issue for CO as the rate constant for the reaction of OH with CO is only weakly pressure dependent. That is, CO loss is efficient throughout the tropical troposphere."

ACPD
7, S1614–S1620, 2007

$>$ p2208/l1: Looking at Figure 2, I see only enhanced variability, but not interannual variations. To support your claim that that variability is due to interannual variations you would need to plot the standard deviation of monthly means - or is this what is shown in Fig2? In any case, a better description of what 'range of observations' means may be appropriate.

This refers to the range of seasonal-averages for each of the five years. We clarified this in the figure caption.

$>$ p2216/l22: A statement like 'more CO crosses the tropopause ... as the ascent rate is higher ... a tape-recorder would exist without changes in tropospheric CO sources' is prone to lead to confusion; and is probably even wrong: A stronger upwelling indeed implies a larger CO mass flux, but it does only indirectly affect CO *mixing ratios*, which is what the term 'tape recorder' is referring to. Upwelling and CO *mixing ratios* are only coupled through the modification of time to reach a certain altitude. Please clarify this in the text.

We removed this statement and have expanded our discussion of the impact of the annual cycle in dynamics in the UT/LS in Section 5.2. We make use of our two new simulations (i.e., tagged CO and uniform tracer) as discussed above.

$>$ p2217/l8: I do not understand what you want to say here. I assume the main upwelling occurs in the tropics, and what you observe in the LMS is outflow from there? Why should then the LMS contribute to the 'tape recorder'?

It doesn't. We say this in the sentence that you are referring too. We include this transport pathway as we find that this is an important cross-tropopause transport pathway for biomass burning pollution, the subject of this paper.

$>$l2221/p4: Note that the Rosenlof (1995) paper refers mainly to the stratosphere, whereas you are probably thinking more of the 150-100hPa layer.

Thank you for pointing this out. We have modified the text accordingly.

ACPD

7, S1614–S1620, 2007

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 2197, 2007.

ACPD
7, S1614–S1620, 2007