Atmos. Chem. Phys. Discuss., 9, C4519–C4530, 2009
www.atmos-chem-phys-discuss.net/9/C4519/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trans-Pacific transport of Asian dust and CO: accumulation of biomass burning CO in the subtropics and dipole structure of transport" *by* J. Nam et al.

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Received and published: 4 September 2009

We appreciate the referees for specific suggestions and comments. Both text and figures has been revised as they suggested for submission to ACP.

Anonymous Referee #1

Specific comment #1. Page 12903, Line 5-7

(Referee) The authors state here that a simulation of aerosols and CO with a 2-month spin-up was conducted for the analysis of the transport events. I am seriously concerned about the 2-month spin-up time for an aerosols and CO simulation. While the

atmospheric lifetime of aerosols is on the order of a month and that of CO is $\sim$ a couple of months, the local lifetime of aerosols (CO) are highly variable, ranging from a few days (weeks) to several months. Spring-time CO at many observation sites in the Pacific is affected by long-range transport of emissions from multiple regions, in particular midlatitude outflow from E. Asia, as well as high-latitude transport that are originated from Europe (Duncan and Logan, 2007) where lifetime of CO can be several months. Therefore, a 2-month spin-up time is not adequate for CO/aerosols to reach steady state. This might significantly affect the simulated CO concentrations, therefore alter the presented results.

(Authors) As the referee indicates in the comment that lifetime of aerosols is on the order of a month, the authors think that the 2-month period is enough for a proper initialization of aerosols. More importantly the referee has a concern that a 2-month spin-up time may not achieve the proper initialization of CO. It is true that the lifetime of CO at midlatitudes in spring can be up to 4 months depending on local conditions including oxidant abundance and meteorology. If this study were to examine CO over "clean" regions, a longer spin-up time would be required. However, our analysis targets large plumes of pollutant exports. The main features of export are not affected as much by the spin-up time as "background" CO. We had carefully looked at the evolution of CO mixing ratio during the two spin-up months and one simulation month. All the CO plumes were subject to fairly rapid dissipation ($\sim$ 1 month) within the Pacific. Therefore, the authors thought that a 2-month spin-up is enough for CO initialization. In order to address the reviewer's comment, we have conducted a 4-month spin-up simulation to see if it makes significant difference in the results of CO plume distributions. Figure 1 compares AOD and CO mixing ratio for the first transport event between using a 2-month (original) and a 4-month spin-up simulation (new). The figure does not show any significant difference in either the AOD or CO distribution of the Asian outflow.

Specific comment #2. Section 4

(Referee) In this section, the authors conducted a sensitivity simulation by increasing biomass burning emissions in the Indochina Peninsula by a factor of 4 (or 8), which yields a better agreement with the MOPITT CO for a single transport event. Thus they conclude that this indicates the GFED biomass burning emissions in the Indochina Peninsula during April 2003 is too low. I am not sure this is a convincing methodology. Here a few of my reasons.

i) A model's ability in accurately reproducing individual long-range transport events of CO depends on many factors, i.e. emissions, transport uncertainties, and OH fields. In particular, lofting of biomass burning emissions in SE Asia is mostly associated with deep convection. An accurate representation of the location or intensity of deep convection remains an active yet challenging research topic. An important question that the authors need to address is how good is the GEOS-4 convection? Was the lifting mechanism for these events well captured by the driving met fields?

ii) While from an averaged perspective, biomass burning effluents from Southeast Asia might occur at lower latitudes than Asian anthropogenic pollution, the transport latitude of an individual biomass burning transpacific transport event can occur in a vast range of latitude band. There a relatively low transport latitude alone does not indicate the bias must be caused by an underestimate in biomass burning emission from SE Asia. Many previous studies have shown that the Asian anthropogenic emissions and biomass burning from SE Asia are commonly mixed in the outflow (e.g. Carmichael et al., 2003; Ma et al., 2003; Bey et al., 2001). Have the authors tried to increase the Asian anthropogenic emissions, which might be a possible alternative candidate in accounting form the model bias?

iii) Figure 3 shows extensive biomass burning in SE China at the same time. I would speculate that if the emissions from SE China were underestimated in the GFED inventory, it would have a similar impact. Have the authors tried to increase the biomass burning emissions in SE China and see how does that affect CO in the Pacific?

I would suggest the authors take a more in-depth approach to investigate the causes of

this model bias, by using tools such as back-trajectories, the GEOS-Chem tagged CO simulation to examine what was the origin of the plumes, its lofting mechanism, was transport too diffusive, etc. In addition to Guam, there are more surface observation sites in the Pacific that can be used to help this analysis. Also, I don't really follow the idea of presenting a 4-day averaged field. This makes it difficult to figure out the origin and pathway of a transport event.

(Authors) We would have done what the reviewer suggested if the goal of this paper is to constrain the sources of biomass burning emissions in May. It is not our goal in part because of large uncertainties in the (a priori) GFED inventory. If we believe that the spatial distribution of biomass burning is correct (not the magnitudes), the major burning areas are over Siberia and Burma, not over SE China. From our model simulations, it is clear that the emission increase needs to come from Burma without resorting to tagged-tracer simulations. However, if the GFED distribution of large fires is incorrect, tagged tracer simulations would not pinpoint the location of the fires responsible for subtropical CO enhancements. Emissions in the southern latitudes would probably have a similar effect. Therefore, we did not emphasize on the significance of potentially large underestimation in Burma in the GFED inventory. Instead, we state in the abstract and conclusions that "Southeast Asian biomass burning emissions" are the reason for the accumulation of subtropical CO. We think that this result is robust.

i) Deep convection is indeed a source of error for global model simulations. For dust, we tested its impact on transport pattern through sensitivity simulations (p. 12910, line 3) and it did not play much role in transport pathway. The description of impact of deep convection has been rephrased, as suggested by the referee #2. For CO, the role of convection is not as significant for CO columns as for the surface mixing ratios. We showed in the paper that both surface and column measurements indicate the effects of biomass burning CO accumulation in the subtropics. Furthermore, the transport pathway of anthropogenic CO from China agrees well with MODIS AOD observations. If convection were dominant in the transport pathway, most aerosols from East China would be scavenged. Perhaps the reason that we did not find convection to be that important is that we selected a period when aerosol AOD enhancements in outflow regions are very large (we note this point in the paper now).

ii) Anthropogenic emissions from SE China release both sulfate (the second most significant contributor to AOD, p. 12908, line 1-4) and CO. We only find enhancements in CO not in aerosol AOD. Further, the CO accumulation weakens rapidly from the first to the latter events. Anthropogenic emissions do not change that rapidly, in contrast to the large decrease of biomass burning emissions from April to May. So we do not believe that anthropogenic emissions are the reason.

iii) Please see the main response. The description of Figure 3 is wrong. Figure 3 shows high surface CO concentration in SE China, mostly from anthropogenic emissions not biomass burning emissions. The whole discussion (p. 12906, lines 11-24) and Figure 3 was cut as suggested by the referee #2. To clarify this point, we include the GFED emission distributions in the supplement (Fig. S1).

Specific comment #3. Section 5-6

(Referee) The details of the comparison between the GEOS-Chem AOD and MODIS AOD were not explained clearly in the text. MODIS usually does not report data under cloudy conditions, while a lot of the Asian outflow happens in the cloudy sector of frontal systems. Did the authors filter the model appropriately for a meaningful comparison between the simulated and observed AOD? It would be necessary to compare daily maps of MODIS AOD with GEOS-Chem AOD to see if the 5-8 May dust transport event happens in the cloudy sector of a frontal system to understand if it is indeed due to model overestimate or it is just that no simultaneous MODIS retrievals were available at the corresponding location. Showing a 4-day average of wind vectors (Figure 9) is not an appropriate way to explain what meteorological systems are involved during this transport episode because this does not offer accurate information of the relative position of the plumes with respect to the pressure systems.

(Authors) As suggested by the referee, the authors have updated the Figure 1 using just GEOS-Chem AOD results corresponding to the cloud-free MODIS observations. MODIS AOD has a sparse coverage due to cloud interference, therefore we compared 4-day average values for transport patterns. Figure 9 shows the overall pattern of wind for the last 4 days of the first event, for consistency with other figures on transport pattern in the manuscript. In the Supplement, wind fields for the first 2 day of the event are shown.

Specific comment #4. Figure qualities

(Referee) Figures 1, 2, 5, 6, 7, and 9 seem to have a resolution problem when they are inserted into the manuscript, which impairs their clarity. Some of the figures were too small, which made it difficult to confirm what's discussed in the manuscript with what's shown in the figures.

(Authors) The authors have inserted the figures with a better resolution. Thank you for pointing this out.

Minor comments:

1. Page 12900, line 24-26

(Referee) Trans-Pacific transport has been well-studied in the past decade and many previous studies showed that trans-Pacific transport has an episodic nature and occur more frequently than once a year. For example, Yienger et al. (2000) shows that 3-5 Asian plumes impact the west coast of the United States' boundary layer between February and May. Liang et al. (2004) suggests that long-range transport of CO from Asia to the northeastern North Pacific region occur year-round every 10, 15, and 30 days in the upper, middle and lower troposphere. None of the three papers cited here by the authors claim that these transport events occur on average only once a year. Table 7 in Jaffe et al. (2003) lists ~2-3 transport events identified during spring 1999 and 2001. The relatively small number of events in the earlier years is a result of limited observations available when long-range trans-Pacific transport was not an active research topic. Heald et al. (2006) identified 4 aerosol transport events in a single spring.

(Authors) The authors have carefully read the references above and have made edits on the manuscript to clarify the statement. Transport of aerosols vs. CO and continuous vs. episodic nature has been distinguished and explicitly explained in the revision.

2. Page 12902, line 15-16

(Referee) How do you reach the conclusion that may 2003 was the period of largest enhancements of trans-Pacific aerosols as well as CO? This was not clearly explained in the text. What was the magnitude of enhancements with respect to the background during this month? What was the average enhancement level? Please give quantitative information

(Authors) Through visual inspection of 7-year MODIS AOD, the authors identified the period of largest trans-Pacific transport (p. 12902, lines 11-12). Visually it is quite clear. We clarify in the paper that it is through visual inspection. We want to identify large events to minimize satellite errors. Quantitative analysis of the relative significance of the enhancement events is not the subject of this paper.

3. Title and Page 12910

(Referee) The importance of the dipolar structure in the pressure/wind fields in the eastern Pacific in regulating the trans-Pacific transport has been noted in several previous studies, Holzer et al. (2005), Liang et al. (2005), and Liu et al. (2005), which should be properly accredited in this study.

(Authors) The references have been accredited in the manuscript.

Anonymous Referee #2

Specific comment:

p. 12903, lines 13-19.

(Referee) The authors should refer to the paper by Fairlie et al. (2007) who describe the implementation of both GOCART (Ginoux et al., 2001) and DEAD (Zender et al., 2003) dust mobilization schemes in GEOS-Chem, compare and contrast the dust emissions distributions using the two schemes separately, apply the model to transpacific transport of dust in 2001.

(Authors) The paragraph has been revised to refer to the reference suggest by the referee.

p. 12906, lines 11-24.

(Referee) I think this whole discussion and comparison to 2006 (and Fig. 3) could be cut. I don't see it as necessary to the theme of the paper, which is explaining differences in observed transport in May 2003. Go straight into showing Fig. 4 to illustrate the improvement with adjusted CO emissions.

(Authors) The paragraph and figure are removed.

p. 12908, line 14.

(Referee) The authors should make clear when they use the DEAD dust scheme (Zender et al., 2003) if they are using the seasonally dependent DEAD source function. The default in GEOS-Chem is to use the GOCART (time-dependent) source function even when the DEAD mobilization formulation is used (Fairlie et al., 2007). If the source functions used are the same (and I suspect they are), then differences between simulations using the DEAD and GOCART schemes will be primarily a matter of magnitude and dust size distribution; the distribution of potential dust sources will be the same. In which case, it should not be necessary to show results from both schemes.

(Authors) The paragraph has been revised as suggested by the referee and the original Figure 1 in the Supplement has been cut.

p. 12910, lines 1-2.

(Referee) It's hard to understand why the dust transport is insensitive to suppressing vertical transport, since clearly it is vertical transport that is responsible for lofting the dust to high altitudes, and the authors note differential advection with altitude in the Pacific high circulation (p. 12910, line 17). If the dust is veered too far south, this suggests that it is transported at too high an altitude, and should respond to changes in vertical advection.

(Authors) Vertical transport is clearly responsible for lifting the dust to high altitudes and different altitude of transport can lead to different horizontal transport pathways, if horizontal wind is sharply different over altitudes. However, sensitivity simulations show that horizontal transport pattern is insensitive to both vertical transport (within a factor of 2) and convection. For vertical transport, since the main sources of dust is at a distance from East China, a smaller lifting rate by a factor of 2 is not enough to change the transport pathway. We noted that horizontal transport could be the error source as the reviewer indicated. The point is now emphasized.

p. 12910, line 3.

(Referee) The authors mention a "third sensitivity experiment," to study the impact of convection, but merely say they don't expect deep convection over arid source regions. This sounds like an argument for discounting convection without having tested it. Did they suppress convection in the model, or not?

(Authors) The authors did a third sensitivity simulations of convection suppression. As indicated in the manuscript, it didn't play a significant role in lifting dust. The paragraph has been modified to clarify.

Main conclusion

(Referee) I find the argument that biases in the dust transport are associated with inaccuracies in the GEOS-4 transport wind field much more persuasive than that this is

due to inadequate dust source locations. I'm certain the dust sources are not perfect by any means, but I'm not persuaded that missing source locations have a "magnitude ... comparable to those from the Taklimakan and Gobi deserts." (p. 12911, lines 15-16). The GOCART source function was updated in GEOS-Chem, since the ACE-Asia study by Chin et al. (2003), to reflect latest Chinese desert maps. This is certainly true for the version v7.03.06 of the model the authors are using. Moreover, the additional sources introduced by Chin et al. (2003) were focused on resolving inadequate simulation of boundary-layer outflow, rather than high-altitude outflow, which the current study shows. So I don't think the argument emphasizing inadequate sources holds in this case. The authors point to differential advection in the vertical associated with the Pacific high circulation, with more pronounced southward displacement at higher altitudes. I suspect that the altitude at which the dust is being transported may be the issue, which points again to the GEOS-4 wind fields, both for transpacific transport and for vertical lofting closer to source regions (see comment on suppressing vertical advection).

(Authors) The authors agree with the referee suggestion. The manuscript has been modified to reflect this point.

Detailed

p. 12900, line 24 – p. 12901, line 5.

(Referee) This is a bit vague. If you mean that transpacific transport occur preferentially in spring, then say so. If there's an increase in background concentrations due to transpacific transport, that suggests the transport is continuous rather than 'once a year'

(Authors) The paragraph has been revised as suggested by the referee.

p. 12901, line 17.

(Referee) The authors say they make use of 7 years of MODIS data, but discuss only

1 month. 7 years of MODIS data doesn't seem that relevant.

(Authors) The sentence has been rephrased in the manuscript.

p. 12908, Fig. 6 and 8.

(Referee) I think that aerosol extinction would seem a more natural quantity to show than "AOD density". Alternatively, the authors could show the aerosols as a mixing ratio.

(Authors) Figures 6 and 8 have been revised in the manuscript, so they show aerosol mixing ratios.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12899, 2009.

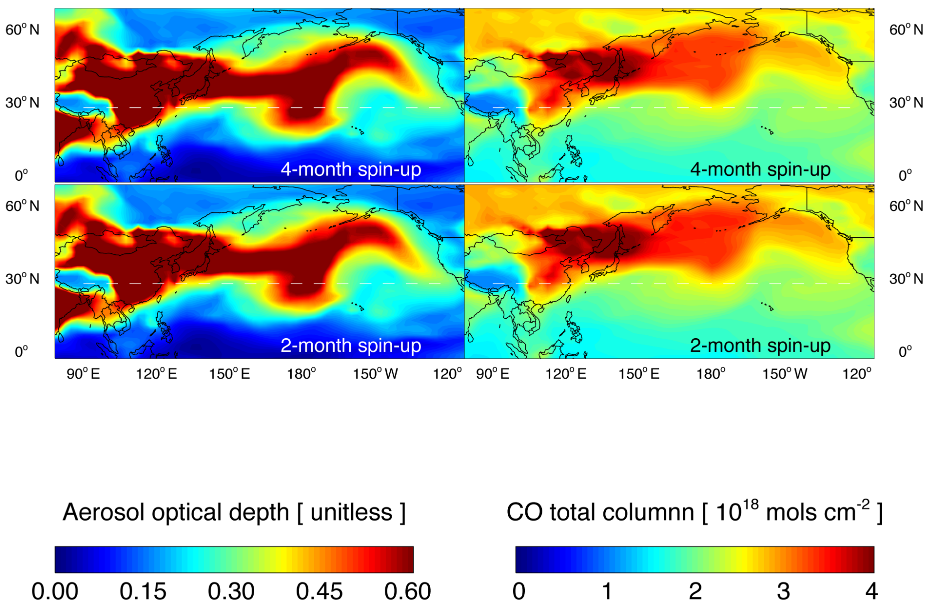

Fig. 1. AOD and CO total columns over the Pacific during 5∼8 May 2003 simulated using spin-up periods of 2 or 4 months by GEOS-Chem.