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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source contributions to Northern Hemisphere CO and black carbon during spring and summer 2008 from POLARCAT and START08/preHIPPO observations and MOZART-4" *by* S. Tilmes et al.

Anonymous Referee #2

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This is not an easy paper to review carefully. This paper compares MOZART-4 simulations of CO and BC to the observations from 6 aircraft campaigns and a satellite instrument (MOPITT). The simulations with MOZART-4 tagged tracers and FLEXPART are used to identify source/region contributions to CO and BC. Many results are presented, but the modeling problems found depend on the source type, location, and season. After reading the paper, one is left searching for the significance of the results. These problems in the MOZART-4 model do not lend themselves to either clear improvements in the model or a better understanding of the atmospheric processes.

If the paper focused on how changes made to the model may have improved model simulations, it would be more useful.

It is pretty clear that BC simulations are not very good compared to the observations. One problem discussed is that wet scavenging of Chinese BC may be too high. I would strongly suggest that the authors do a sensitive simulation (for example, making BC much more hygroscopic) and show where and how it improves BC simulations. Since the model cannot simulate BC observations well, it would be better to simply remove the discussion of BC contributions from different sources in section 4.

One of the problems with CO simulations is that when adding tagged CO tracers ("anthropogenic" and biomass burning; by the way, biomass burning CO is usually considered to be anthropogenic), it is about 50% of CO in winter and 30% of CO in summer. The fraction by each source is often < 10% of the total CO. In comparison, the fraction not tracked by tagged tracers is much larger. Some of the model errors can certainly come from erroneous simulations of that fraction of CO. This error is ignored. There are other possibilities including transport, chemical yield of CO, and fire emission distribution. I do not get a sense that all possible errors were investigated in order to find some common themes among the model comparison problems.

Other comments.

1. Delete Figure 2. It does not show more information than described in the text.

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2. L15-20. The statement that " but this does not change the partitioning of the total sources" is incorrect. CO from CH4 and isoprene, > 50% of total CO, is not "anthropogenic". In addition, the CO yield from anthropogenic VOCs varies with many factors such as NOx, light, T, water vapor. It is not a constant.

3. L23-24, are "lower" and "higher" relative to the values by Fisher et al.?

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4. L5-7, one would think that the simulation of surface emissions only is very different from evenly distributing the emissions from surface to 6 km. Figure 9, for example, clearly shows the vertical dependence of CO from Canada/Alaska fires, which would be wiped out if the emissions have not altitude dependence. Can the authors do a sensitivity simulation to confirm this statement?

5. L9-15. Delete Figure 3. It does not provide new information.

6. Figure 5 shows that the tagged CO is only 50% of total CO. The contribution from China, which is the largest, is < 15% in the second column. I think it is difficult to say that the underestimate/overestimate of the model CO is from one or some of the anthropogenic sources.

P. 5947

7. L19. Transport could also be a problem (too much from a high emission region).

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8. L1-5. Transport could be a problem. Could there also be a problem of CO yield from VOC oxidation in the model? (The yield is too low).

9. L9-10. This statement contradicts L16-17 on P. 5947, which stated " Since fire emissions are not a very important contributor for ARCTAS CO and BC measurements in early April".

10. L26. The model result here does not seem to agree with the observations. The observed BC peaks are not at 9 km. Fire tagged BC peaks at 9 km or 4 km in the 2nd column. The observed peak is at 5-6 km.

P. 5949 11. L4-6. If 10% of CO underestimation is from anthropogenic sources and the total is 15%, the underestimate from fires is only 5%. Is this right? In the paper (conclusions), it sounded like the biomass burning emission is underestimated much more than anthropogenic emissions.

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12. L10, "50%" is more like 70%.

13. L11. Increasing scavenging can help the simulation above 8 km. It will also make the model underestimate at 3-6 km much worse. Model simulations need to show what happens if BC scavenging is increased.

14. L20. FLEXPART shows that CO at 6 km is from Europe. Figure 5 shows that CO from Europe is at 3-5 km. It looks like a clear model transport problem.

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15. L1. It is not just an emission magnitude problem. The altitude range from MOZART (2-5 km) of fire CO is much narrower than FLEXPART (1-8 km). It looks like a problem either in transport or emission distribution.

16. L3. What are "high altitudes"? The first column shows model is ok. The second column shows that the model is higher than observations. The third and fourth columns have no data above 7 km.

17. L11-12. Is the comparison in Figure 7 similar to Figure 5? The difference in the 1st and 2nd columns of Figure 5 seems much less than Figure 7. The 3rd and 4th columns do not have enough data. From the previous discussion, the underestimate of 10% of CO is from anthropogenic sources and 5% is from fires. Could the MOPITT comparison be explained by a problem in anthropogenic emissions? What are the reasons for much higher CO in MOZART in the subtropical(?) Pacific?

18. L26-28. The sum of all tagged CO is < 20 ppb. The total CO is 100 ppb. To say a source from a region dominates seems to overstretch its significance.

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19. L9. A model simulation needs to show how increasing scavenging helps.

20. Figure 9. The sum of tagged CO is 20-30 ppb out of a total CO of >100 ppb. All anthropogenic emissions have to be tripled to make up for a difference of 40 ppb.

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21. L2-5. Unlike the FLEXPART result, the model result in Figure 9 doesn't show more East Siberian CO at 8 km than other altitudes. Is this a model transport problem?

22. L10-15. Figure 9 shows Canadian CO is only 2-3 ppb at > 5 km. Increasing the Canadian CO emission by any reasonable amount is not going to help solve the problem.

23. L24-30. The anthropogenic and fire sources (tagged) are too small to make up for the large difference.

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24. L14-17. Where is the evidence for the overestimates of Asian fires? In Figure 12, if fire emissions are overestimated over the Asian continent, why is the model too low over the western Pacific? The outflow region should have high CO from the overestimated emissions.

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