Atmos. Chem. Phys. Discuss., 11, C1266–C1273, 2011
www.atmos-chem-phys-discuss.net/11/C1266/2011/

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 #1

Received and published: 28 March 2011

Tilmes et al. 2011 presented a comparison of simulated CO and Black Carbon from the MOZART-4 model against aircraft measurements obtained during several aircraft campaigns, including NASA ARCTAS, NOAA ARCPAC, POLARCAT-France, etc. By tagging CO and BC from various sources, they also estimated the relative contribution from various sources and regions to tropospheric CO and BC in the N. Hemisphere. Overall I found this paper to be a redundantly long paper that adds little new scientific findings to our existing knowledge of long-range transport into the Arctic. I have three major concerns:

i) Lack of significant new scientific findings. Many of the observations used in this paper and the associated CO and BC source attribution have already been discussed in other papers (e.g. Fisher et al. 2010 for ARCTAS and ARCPAC, Pfister et al. 2010 for ARCTAS-CARB, Paris et al. 2009 for YAK-AEROSIB). If the focus of this paper is about source attribution of long-range transport of pollution into the Arctic, what are the new findings this study adds to what have already been revealed by Fisher et al. (2010) and Paris et al. (2009) as well as many previous papers, e.g. Stohl et al. (2006), Shindell et al. (2008)? In their introduction, the authors explained that they aim to present a more comprehensive study by including measurements from preHIPPO and START08 to quantity the sources of CO in the entire NH. First all, the preHIPPO and START08 missions only cover the N. American region, therefore do not represent the entire NH. Source attribution of CO can be very different in Asia or Europe from that in N. America. Secondly, after the extensive studies conducted for the NOAA ITCT-2K2, NASA INTEX-A and INTEX-B missions, what are the new findings the preHIPPO/START08 measurements offer on CO/BC sources that were not learned from the previous campaigns?

ii) From what presented in this paper, there still exist significant model biases in accurately reproducing the observed CO and BC concentrations. The authors attribute these model biases to underestimate in anthropogenic emissions and Canadian fire emissions and overestimate in Siberian fire emissions (for CO) and underestimate of fire emissions. The model bias for BC seems particularly alarming and can only be partially explained by errors in wet scavenging, with the remaining large model error not account for. Even when the simulated total CO agrees well with the observed CO during START08, the authors suggest that this good agreement is “a result of two compensating shortcomings of the model: the overestimation of Asian fires and the underestimate of anthropogenic emission in mid-latitudes. I would encourage the authors to address this issue by obtaining an improve emission estimate based on what revealed from the model-observation comparison and rerun the simulation with the improved emission estimate. Until these existing issues with the emissions (scavenging

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as well) are resolved, what's the meaning of discussing the source attribution using only the significantly biased model results?

iii) Identify the source of model biases. Throughout the text, the authors attribute all identified model vs. observation differences in CO to underestimate and/or overestimate in emissions. However, there are two additional factors which can contribute significantly to simulated CO, uncertainties in transport and model OH fields. An accurate representation of OH is essential in CO simulation in two ways. It can introduce a bias in CO sources through underestimate (and/or overestimate) of CO produced from CH4 oxidation. High (low) OH can also lead to too much (little) destruction of CO, therefore a bias in CO sink. How good is the OH field simulated by MOZART-4? What's the implied CH3CCl3 lifetime? There are also OH measurements available from ARCTAS and possibly other campaigns. How are the MOZART OH compared to these observations? In addition, how reliable is the model transport, especially deep convection? These questions need to be addressed before attributing all model biases to uncertainties in emissions.

iv) I have also a long list of minor concerns which I feel impair the clarity and quality of the manuscript. The particular worrisome issue is that a lot of the conclusions in the text on possible causes of model biases were drawn without clear and sound evidence.

1. Page 5938, lines 8-10. I don't see how does CO transported into the Arctic influence ecosystems?

2. Page 5938, lines 13-15. How does O3 (therefore CO) influence climate? Not everyone knows this. Please clarify?

3. Page 5939, line 23. "entire NH"- The measurements only covers the Polar region and N. America, not the entire NH.

4. Page 5939, lines 26-29. While CO has commonly been used as a tracer of pollution transport, BC is not a very good indicator of transport. Its lifetime is short and is

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efficiently removed by wet scavenging and sedimentation.

5. Section 2.1. Need to add a brief description of the instruments used to measure CO during various missions.

6. Page 5943, lines 27-28. "... is often in agreement with the origin of ..." How often is often? Please quantify. Also I would suggest the authors to include a comparison of the source regions identified by FLEXPART with the model tag tracers. How good does the two agree with each other? This can help significantly in increasing the robustness in both.

7. Page 5945, lines 14-17. What are the scale factors used to account for CO produced from the non-methane hydrocarbons?

8. Page 5946, lines 15-17. Does the model output interpolated spatially or temporally or both? Please clarify.

9. Page 5947, lines 18-19. "a large part of the differences between the model and observations at high latitudes in spring can be attributed to anthropogenic emissions" – As I have already discussed in above, how do you know the differences is due to emissions? It could be due to model biases in OH.

10. Page 5947, lines 27-28. "A significant influence of Siberian fire emissions is not simulated by the model ... during POLARCAT-France." How do you know it is Siberian fire emission? I can not even find in these two panels a line for the Siberian fires.

11. Page 5948, lines 1-5. "Difference of more than 10% between modeled and observed CO... either as a result of the impact of Siberian fire emissions ... or due to an additional underestimation of anthropogenic emissions coming from Europe.." Again, this is merely speculation. Any supporting evidence? I don't see a line represent Siberian fires. The dominant impact of European emission does not exclusively lead to the underestimate of European anthropogenic emission as a source of model bias. In fact, if the underestimate of European anthropogenic emission was the reason, one

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would expect to see the maximum model-observation difference in below 4 km where the European contribution maximizes, which is clearly not the case (the difference remains relatively constant between 0-8 km).

12. Page 5949, lines 10-11. What are the evidences support your assumption of an underestimate of the contribution of anthropogenic emission of ∼10%? On Page 5928, lines 1-5, the authors also suggest the 10% difference could be due to either anthropogenic emission or Siberian fire emissions.

13. Page 5949, lines 6-15. The simulated BC during ARCTAS:04/12-17 and ARCPAC are 4-6 times the observed values and the vertical profiles looks nothing like the observed profiles? Why the model fail to reproduce the very high BC levels between 2-8 km? Any explanation on possible causes. Are there any issues with emission? Is it because of transport errors, therefore not efficient lofting? If the main focus is about understanding the source attribution, it is important to address all the model biases. Also I don't see how the results from Matsui et al. (2011) are tied to the model biases shown here.

14. Page 5949, lines 21-22. Here the authors suggest that airmasses from Europe impact altitudes of 3-8 km for CO and BC. This is in conflict with the results shown in Figure5, with maximum European emission near the surface. Why the difference? 15. Page 5949, lines 28-29. I am very confused by this sentence. Which figure and which CO profile are you referring to?

16. Page 5950, line 5 & Figure 7. The enhanced CO in East Siberia is not clear to me. There's not much data available and may be you want to change the color scale to make it more identifiable.

17. Page 5950, lines 6-7. What are meteorological conditions? Please explain with more details.

18. Page 5950, lines 11-13. I am not convinced that the underestimate of CO at 400

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hPa implies that the Siberia fire emission is underestimated. Enhanced CO values in East Siberia as observed by MOPITT does not necessarily mean the underestimate in CO over Alaska at the same time period has to be due to the underestimate in East Siberia biomass burning emission.

19. Page 5952, lines 10-12. "Enhanced airmasses from Alaska and Canada" – What enhanced airmasess? And which figure you are referring to?

20. Page 5952, lines 12-14. ". . . the result of an underestimate of the impact of Canadian fire emissions on higher altitudes". What are the facts used to support the above conclusion? In fact, by just looking at the figures, the model seem to significantly underestimate the peak at 6 km which is associated with the E-Siberia fire emission, which may imply an underestimate of the contribution from the fire in E-Siberia. In addition, the current contribution of Canadian fire emission is $\sim$1-2 ppbv at > 5 km. Is it reasonable to suggest an underestimate of Canadian fire emission can explain the 30-50 ppbv difference in total CO between the model and observation?

21. Page 5952, lines 19-24. How do you know the enhanced CO over the western Pacific is associated with E-Siberian fires? It could be due to outflow of Chinese anthropogenic emissions which are favored in summer.

22. Page 5952, lines 27-29. Figure 9 shows that with the current emission set up, the U.S. anthropogenic emission and E. Siberian contributes $\sim$10 ppbv to total CO. To account for the 30% difference (30-40 ppbv), this implies both have to increase 3-4 times to match the observed CO. The simulated CO for YAK-AEROSIB, GRACE, FRANCE (Figure 9), which is already too high in general, would also be increased by an additional 30-40 ppbv (similar contribution from the two sources in these regions). This to me means the speculated reason is not the real cause of the model bias.

23. Page 5953, lines 19-20. What do you mean by European fire? It was not mentioned in previous text.

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24. Page 5953, line 25. “Uncertainties between model and observations, . . .” Not sure what uncertainties are you talking about.

25. Page 5954, lines 15-16. “The high CO values at high altitudes (above 12 km) are very likely a result of the Indian monsoon that occurs at around 30N.” How does the Indian monsoon around 30N spread its influence to high altitude CO in the mid and high latitudes? Please explain in detail.

26. Page 5954, lines 21. How do you know its “forest fires come from China and Southeast Asia”? It can be other biomass burning sources, for example agricultural burning?

27. Figure 5 (similarly figures 9 and 13). The authors may want to explore a clearer and more efficient way of presenting the model results. This figure contains way too many lines. In the top and 3rd rows, why show the 5 and 95 percentile lines if i) they are not discussed in the text, ii) they really make the comparison hard to see. Also I find it difficult, in several cases, to tell apart or find the lines for some of tagged tracers, e.g. U.S. vs India, SE Asia anthropogenic emission vs fire emissions.

28. Inadequate reference of previous literature. The source attribution of CO (and BC) in the Arctic has been extensively studied in previous studies. To name a few, Echhardt et al., 2003; Koch and Hansen, 2005; Stohl et al., 2006; Shindell et al., 2008. There are also many related papers looking at source attribution of CO (and BC) in North America using ITCT2K2, INTEX-A, INTEX-B data. The results from these earlier studies are highly relevant to this analysis, in particular section 4, but none were cited. In fact, section 4 did cite a single paper. I strongly feel that this is a clear sample of inadequate reference of previous literature.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 5935, 2011.