Atmos. Chem. Phys. Discuss., 11, C4602–C4629, 2011
www.atmos-chem-phys-discuss.net/11/C4602/2011/

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.

S. Tilmes et al.

tilmes@ucar.edu

Received and published: 9 June 2011

General Response

We thank both reviewers for a detailed review and important suggestions that have helped to significantly improve the paper. Based on the review comments we performed the following changes to the manuscript. A detailed response to each point raised by the reviewers can be found below.

We added a new Chapter 3 to describe the MOZART-4 model and its general perfor-

mance. We moved Section 2.2 to Section 3.3, which allows us to introduce the general description of the performance of MOZART first and afterward the comparison to results using FLEXPART trajectories. Consequently, Figure 3 is now Figure 2.

Section 3.2 is added 'MOZART-4 representation of source regions in comparison to MOPITT' to quantify the general performance of the model and therefore the representation of emissions in the model. Within this Section, we added two new Figures, Figure 3 and Figure 4. In Section 3, we also added a new Table 3: comparing MOZART-4 OH with the OH climatology from Spivakovsky et al., 2000. On the other hand we removed old Figures 7,11 and 12 (the 10-day average comparison between MOZART and MOPITT.

The old Figure 2 is now Figure 5.

We added an additional panel to the new Figure 5 (old Figure 2), comparing CO tags as derived by MOZART-4 with results derived using FLEXPART trajectories.

Old Figure 4 is now Figure 6.

Old Figure 5 is now Figure 7.

Figure 7 was changed: Row 2 and 4: we show relative contributions of CO and BC tags to the total amount of tagged (directly emitted) CO instead of absolute values. W/E Siberian tags are combined and colors are changed. We also illustrate the 5^{th} and 95^{th} percentile as a shaded area (for the observations) to improve the readability of the Figure.

Old Figure 6 is now Figure 8

Figure 8: we removed the third (right) row, showing CO tags for Europe.

Figure 9: changed as described for new Figure 7

We switched Section 3.3 and 3.4 (new Sections 4.4. and 4.3) to discuss results in high latitudes first and then discuss comparisons in mid latitudes.

ACPD

11, C4602–C4629, 2011

Figure 10: changed, as described for Figure 8

Old Figure 8 is now Figure 11: changed as described for new Figure 7

Old Figure 13 is now Figure 12: changed as described for new Figure 7

Older Figures 14,15 and 16 are now Figures 13,14 and 15.

Changes to Figure 13: we show relative CO contributions with regard to the total amount of tagged CO instead of the relative CO contribution with regard to total CO.

Response to Reviewer1:

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

ACPD

11, C4602–C4629, 2011

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?

We thank the reviewer for those comments. The goal and scientific findings of the paper as presented in the submitted version were indeed not clearly stated and we improved this in the new version of the manuscript, as outlined below. We also agree that many observations have already been discussed in earlier papers. Further, the source contributions of long-range transport of pollution to the Arctic have been discussed in earlier studies. However, as Shindell et al., 2006 and 2008, pointed out that there are still large uncertainties in models to simulated CO and BC (Koch et al, 2009, Shindell et al., 2009). Further observations to evaluate chemistry climate models are limited and shortcomings of models are not identified.

The goal of this study is to simulate BC and CO during POLARCAT 2008 in spring and summer using MOZART-4 and to identify shortcomings of the model. Model results are evaluated in detail using 5 POLARCAT aircraft missions as well as the START08/preHIPPO mission. Using a combined set of aircraft observations provides a good representation of airmass characteristics over the entire polar region and over North America in spring and summer 2008. Further, the different campaigns employed here are influenced by different amounts and types of pollution. The representation of model results was therefore tested for the varying influence of pollution as observed during the different aircraft campaigns and different periods. Further, CO and BC contributions derived in this study are compared with results from earlier studies. Differences between different seasons and reasons of source contributions are discussed.

START08/preHIPPO aircraft observations are included in this study because this campaign did not target any pollution plumes. The model performance for mid-latitudes over the US was investigated to complement other aircraft observations in mid and high latitudes that targeted pollution plumes during the same season and year. Differ-

ACPD
11, C4602–C4629, 2011

ent than using earlier mid-latitude aircraft campaigns, START08/preHIPPO took place during the same year as POLARCAT 2008.

To investigate the presentation of emissions over source regions, we add a comparison between MOPITT satellite data and MOZART-4 at 800 hPa for important source regions, as well as for 400 hPa over Alaska and Canada in the revised version of the manuscript. This allows us to obtain further insights in the representation of simulated CO.

Aircraft data as used in the study can be employed in a similar way to further model comparison activities. This can help to identify sources of shortcomings in different models and eventually improve the prediction of future climate. In the new version of the manuscript, we change the introduction (and other parts of the paper) in a way that the goal of the study, as described above, is more clearly stated.

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

ACPD

11, C4602–C4629, 2011

emission estimate. Until these existing issues with the emissions (scavenging as well) are resolved, what's the meaning of discussing the source attribution using

only the significantly biased model results?

We agree with the reviewer that based on how the results were presented in the submitted manuscript, the reader would come to the conclusions that shortcomings in the model could be improved by changing the amount of emissions, however, we come to the conclusion that to the most part this is not reasonable here, as outlined:

In the revised version of the manuscript we have added a comparison of CO between MOZART-4 and MOPITT over different source regions (Section3.2, Figure 3 in the new manuscript). Since the model represents CO very well for most of the source regions besides Southeast Asia (as addressed with the next comment), we conclude that local emissions are presented reasonably well, within the variability of the observations. Further, CO and BC is simulated reasonable well in mid-latitudes. Base on the location and timing of observations, the underestimation of CO and BC especially in fire plumes at high latitudes are ascribed to the dilution of fine-scale pollution plumes in the coarse grid of the model. There are further uncertainties in comparing model results on the flight track.

Therefore, the adjustment of emissions as done for example in Fisher et al, 2010, would not be reasonable in this study. The improvement of scavenging in the model is not straightforward and is a task for future model development.

Since the comparison between MOZART-4 and MOPITT shows a reasonable agreement over Alaska-Canada, large uncertainties between model and aircraft data might be not very significant, since large pollution plumes were targeted, that are not representative of the entire region. A discussion about source attributions of CO and BC is therefore reasonable.

iii) Identify the source of model biases. Throughout the text, the authors attribute all

ACPD

11, C4602–C4629, 2011

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.

In the revised version of the manuscript we use the OH climatology from Spivakovsky et al., 2000, to compare to OH distribution of the model. MOZART-4 underestimates OH with a maximum in low latitudes and a minimum in high latitudes, although with discrepancies more significant in summer than in spring at high northern latitudes below 500hPa (see new Table 3). Since simulated CO is in good agreement with MOPITT observations over source regions in mid- and high latitudes, but CO is overestimated in low latitudes, we assume that only low latitude CO is impacted significantly. The good agreement of CO between MOZART-4 and MOPITT at both 800 and 400 hPa over different regions (only shown for AK-Canada in the revised version of the paper, see Figure 4) indicates a reasonable representation of deep convection. We include a discussion of the described uncertainties, including transport in the model, in the revised version of the paper. We do not calculate the CH3CCl3 lifetime in MOZART-4 (also, it is only a good measure of OH in the Tropical lower troposphere [Lawrence et al., ACP, 2001]). Moreover it has been recently demonstrated that OH inference from methylchloroform were leading to overestimate of OH interannual variability and therefore are not anymore a good target for model OH validation since $\sim$10years (Montska et al., 2011, Science, doi:10.1126/science.1197640). This further justifies the use of a

ACPD

11, C4602–C4629, 2011

comparison with a climatology as done here, vs. comparison with CH3CCl3.

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?

CO is a precursor of ozone, which then can influence the ecosystem. (The introduction is rewritten and the reference to ecosystem has been removed.)

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

O3 is a greenhouse gas and therefore influences climate. We have included the explanation in the introduction. 'CO is an important precursor to ozone due to its influence on the abundance of OH and on the oxidation capacity of the troposphere. Therefore, CO has an influence on the tropospheric ozone budget and indirectly on climate, because ozone is a greenhouse gas.'

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

This is clarified in the introduction of the revised paper: See also the response to general comment i. 'The latter [START)8/preHIPPO] took place for the most part in midlatitudes over North America during the same period as the POLARCAT campaigns and did not target any pollution plumes inthe troposphere. These additional observations complement the POLARCAT observations, due to the different target and location of the campaigns.'

4. Page 5939, lines 26-29. While CO has commonly been used as a tracer of pollution

ACPD

11, C4602–C4629, 2011

transport, BC is not a very good indicator of transport. Its lifetime is short and is efficiently removed by wet scavenging and sedimentation.

We agree and remove this statement.

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

We added the description and references to this Section in the revised version of the manuscript, in Section 2.1, Line 140ff of the revised manuscript: 'During the POLARCAT mission, CO was measured using the following instruments, a Diode Laser In-Situ (DACOM) during ARCTAS with an accuracy of 2% (Sachse et al., 1987), a vacuumUV fluorescence instrument during ARCPAC with an accuracy of 1% (Holloway et al., 2000), an IR absorption gas correlation gas analyser with an accuracy of 5% during both POLARCAT-France and YAK-AEROSIB (Nedelec2003; Paris et al., 2008), and a vacuum ultraviolet (UV) fl uorescence technique using an AEROLASER instrument with and accuracy of 5% (Baehr et al., 2003) during POLARCATGRACE, as described in more detail in Pommier et al. (2010). During START08/Hippo CO was measured using Aircraft laser infrared absorption spectrometer with an accuracy of 2% (Webster et al., 1994; Lopez et al., 2008) as described in Tilmes et al. (2010).'

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.

We added an additional panel to the new Figure 5 and rewrote the text according to the comment and the new panel in Section 3.3. Information from trajectories do not allow to keep track of the amount of pollution that entered traced airmasses, which

ACPD

11, C4602–C4629, 2011

are influenced by various sources, but just of the location of airmasses. Consequently, we do not expect an agreement between identified source region using FLEXPART trajectories and tagged ozone from MOZART-4. However, trajectories are helpful to identify the source of large fire plumes. Indeed, detected fire plumes from East Siberia show enhanced mixing ratios of the CO tags from East Siberia, however CO tags from MOZART-4 are underestimated in this case.

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

The secondary production of CO is kept track of separately, without any source attribution in the model. The tagged CO is only from direct emissions of CO. Fig. 15 shows the contribution of CH4 and NMHC oxidation to CO (the difference between the black line for total CO and the top of the color contours which is the sum of the tags).

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

Both, this is clarified in the new version.

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.

This is discussed in the answer to the general comment III. We change the sentence to:

'a large part of the differences between model and observations at high latitudes in spring can be attributed to other factors, e.g., the underestimation of local anthropogenic emissions over Alaska and Canada, as well as the underestimation of the

ACPD
11, C4602–C4629, 2011

remote influence of CO sources most likely from anthropogenic emissions.'

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.

DeVilliers et al., 2010, have found a large influence of Siberian fire emissions during POLARCAT-FRANCE. MOZART-4 does not simulate a large amount of CO coming from Siberian fires and that is why there is no line representing those emissions. It is very likely that the model underestimates this influence. We have clarified this part in the revised version of the manuscript: 'Different than suggested by Adam De Villiers et al. (2010), no significant influence of Siberian fire emissions is simulated by MOZART-4 for areas investigated during POLARCAT-France. This points to an underestimation of the infl uence of Siberian fi re emissions in Northern Europe in the model.'

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. (as discussed in the previous comment).

We agree with the reviewer and have removed the assumption that European anthropogenic emissions might be underestimated because we do not have any evidence for this.

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

ACPD

11, C4602–C4629, 2011

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).

We agree, that European emissions are likely not the cause for the larger difference between model and observations during POLARCAT-France. Again, we remove this statement.

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.

The assumption was based on the fact that CO was underestimated by 10% in early April mostly as a result of the underestimation of the remote influence of anthropogenic emissions. However, as we have learned from the new comparison between MOPITT and MOZART, this difference disappears after the second week in April. Discrepancies between model and observations are likely caused by the underestimation of CO and BC in fire plumes in high latitudes in the model (as discussed above). We rewrote this paragraph to include the new conclusion.

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

ACPD
11, C4602–C4629, 2011

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.

Besides the underestimation of large pollution plumes form Siberian, due to the dilution of pollution plumes in the model and transport errors, as the case of CO, the model might also underestimate the remote influence of anthropogenic BC emissions.

However, this is an assumption, since there are no observations available to compare to the model. Further, the MOZART-4 largely overestimates BC above 8km by about 1 order of magnitude, which can be ascribed to South Asian (Chinese) anthropogenic emissions by the model tags. These differences are likely caused by an underestimation of wet scavenging of BC in the model and a potential overestimation of BC emissions in China. We do not expect significant issues with emissions or vertical transport, as shown in the case for CO, while comparing to MOPITT satellite data. We remove the reference to Matsui et al., 2011.

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?

We agree with this concern. As explained above, FLEXPART trajectories are not used to identify the contributions from different source regions. Enhanced CO mixing ratios that have their source in Europe can be strongly influenced from Siberian fire emissions, which lead to enhanced CO mixing ratios at 3-8 km. However, the maximum contribution of CO from European source regions is still at the surface. This is confusing and therefore we do not discuss FLEXPART trajectories from Europe in the revised version of the manuscript.

15.

ACPD

11, C4602–C4629, 2011

Page 5949, lines 28-29. I am very confused by this sentence. Which figure and which CO profile are you referring to?

We have removed this sentence.

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.

We have removed this paragraph and Figures 7, 11 and 12, and instead performed a comparison between MOPITT and MOZART over source regions, as described above.

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

This part of the text is removed in the new manuscript.

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

Removed, as discussed above.

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

We have changed this paragraph. The point is that FLEXPART trajectories (old Figure 10) suggest enhanced values of BC and CO have their source in Alaska/Canada.

ACPD

11, C4602–C4629, 2011

However, as discussed above, airmasses are likely strongly influenced by pollution from other source regions, which are underestimated in the model.

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 _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?

We have rewritten this paragraph. Differences between model and observations are more likely a combination of different regions, including the underestimation of Canadian fires in higher altitudes and the underestimation of the remote influence of Siberian pollution. The use of absolute CO model tags is misleading in the earlier version of the paper, because the photochemically produced CO is as important.

'The underestimation of CO above 4 km is likely a combination of different reasons. FLEXPART

trajectories (Figure 10) suggest enhanced BC and CO values above 5 km (up to 220 ppbv for CO)

with source regions in Alaska and Canada. This points to an underestimation of local pollution

ACPD

11, C4602–C4629, 2011

in altitudes above 5 km. However, identified airmasses using FLEXPART trajectories might be also

strongly influenced by pollution from other source regions (as discussed in Section 3.3), which might

be underestimated in MOZART-4.'

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.

We have dropped the comparison between MOPITT and MOZART-4 here as well.

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 _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.

We agree with the review that the argument is misleading and dropped it. In general, in the revised version we are not showing total contributions of CO tags, but relative contributions of the total tagged (directly emitted) CO for each source regions. Values given in the previous version of the manuscript only show tags from direct CO emissions and do not include photo-chemically produced CO in that region. We assume that both scale the same and show relative contributions instead. Further, the large

ACPD

11, C4602–C4629, 2011

underestimation of CO during ARCTAS-B is likely a result of the underestimation of fire plumes as targeted during these flights. However, the emissions over the source regions are not underestimated.

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

This was a typo and "European anthropogenic emissions" was meant instead.

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

Changed to "Differences between model and observations.."

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.

Highest contribution from India occurs at high altitudes over the Arctic because of lofting of airmasses (convection) by the monsoon. In around 30N, CO contributions from India reach up to 40% above 10km (see new Figure 13). We clarified this in the new version of the paper. 'Indian anthropogenic emissions are very strong in summer and contribute up to 40% to the pollution in low latitudes above 12 km. High CO mixing ratios accumulate every summer in the upper troposphere around 30 N as a result of convection in the Asian monsoon (e.g., Dunkerton, 1995). Some of those Indian emissions are transported towards high latitudes and contribute (≈5%) to the entire CO abundance in the Arctic region.'

26. Page 5954, lines 21. How do you know its "forest fires come from China and

ACPD

11, C4602–C4629, 2011

Southeast Asia"? It can be other biomass burning sources, for example agricultural burning?

We changed this to a more general statement.

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.

As discussed above, relative contribution of CO and BC are shown in the new version of the paper. Further, to make the Figures easier to read, we combined the contributions from East and West Siberia and changed the color for India. The 5 and 95 percentiles are not removed. We think, that it is important to also compare the range of the distribution, which are discussed in the paper. To improve the figure, we show the 5^{th} and 95^{th} percentile for the observations as a shaded area.

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

ACPD

11, C4602–C4629, 2011

inadequate reference of previous literature.

We thank the reviewer for pointing us to relevant literature. In the revised version of the manuscript, we add more relevant references and compare the result from this study to those from earlier findings.

Respond to Reviewer2

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.

We agree with the reviewer that the goal of the paper, as well as the results presented in the submitted version of the paper were not clearly stated, please see response to Review 1. We have rewritten the introduction, revised the discussion of the new sections 4 (old section 3) and included a summary of the results based on the comparison between aircraft and MOZART-4 data.

If the paper focused on how changes made to the model may have improved model

simulations, it would be more useful.

Based on the analysis, and stated in the revised version of the manuscript, most sig-

ACPD

11, C4602–C4629, 2011

nificant shortcomings can be ascribed to the underestimation of BC and CO in pollution plumes in high latitudes, as a result of the dilution of fine-scale fire plumes in the coarse model grid and possible uncertainties to compare the model with data on the flight track. Therefore, the adjustment of emissions as done for example in Fisher et al, 2010, is not reasonable in this study for the most part (as also described in the Response to Review 1). With regard to CO, the study shown that the performance of the model is reasonable in mid and high latitudes in comparison to MOPITT satellite observations. We have pointed this out in adding a new Section 3.2 'MOZART-4 representation of source regions in comparison to MOPITT' to the paper.

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.

The improvement of scavenging in the model is not straightforward and is a task for future model development. One goal of this study was to identify shortcomings of the model. We do not think that just changing BC to be more hygroscopic will resolve the problem, but we need more sophisticated changes that are beyond the scope of this paper. However, we strongly agree with the reviewer that this is an important research topic for future studies.

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.

We agree that shortcomings in simulating BC in MOZART are large. In section 4, some reasons for the shortcomings could be for the most part identified, as the dilution of fire plumes in the model and the lack of wet scavenging. We note that largest discrepancies often occur for aircraft campaigns that targeted fire plumes. Differences might be

ACPD

11, C4602–C4629, 2011

smaller if considering a larger region, as shown for CO. We leave the discussion of BC contributions in the paper but add information about possible shortcomings: 'Since the MOZART-4 significantly underestimates BC values in mid and high latitudes below 8 km in spring as a result of an underestimation of fi re plumes that reach high latitudes, the BC contribution from East and West Siberia is likely underestimated in Figure 14. On the other hand, the contribution from Asian anthropogenic BC emissions in higher altitudes is likely overestimated in the model.'

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.

In MOZART-4, we separate between two types of emissions, those from anthropogenic sources including fossil fuel and those from open burning, including both wildfires and agricultural burning. For CO tags in MOZART-4 only CO from direct emissions is considered. In the revised version of the manuscript, we are not showing total contributions of CO tags, but only the relative contributions of the directly emitted CO for each source region. We assume that direct emissions and photochemically produced CO scale similarly. Only small errors might be introduced in the assignment of sources to the total CO budget (as mentioned in the new version of the manuscript). The comparison between MOPITT and MOZART-4 over different source regions indicates a reasonable assumption of CO emissions.

There are other possibilities including transport, chemical yield of CO, and fire emission distribution.

ACPD

11, C4602–C4629, 2011

I do not get a sense that all possible errors were investigated in order to find some common themes among the model comparison problems.

In the revised version of the manuscript, we add a comparison between MOPITT and MOZART-4 over different source regions that indicates a reasonable assumption of CO emissions. Further, the OH distribution of the model is evaluated and potential shortcomings of horizontal and vertical transport are discussed (see response to Reviewer 1). The possible errors in the model are pointed out more clearly in the revised version of the paper.

Other comments.

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

We added a new panel showing relative contributions of CO tags from different regions to the flight path and revise the text (see new Figure 5). This comparison is important to interpret results from MOZART-4 CO tags and source regions derived using FLEXPART trajectories. This comparison was also suggested by Reviewer 1.

$P5945$

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.

Fig. 5 of Pfister et al. (JGR, 2008) shows that the contribution of isoprene oxidation to total CO is less than 10% and methane oxidation is about 20% of CO over China. The remainder of the chemically produced CO (approx. 30% of CO) is likely from oxidation of anthropogenic and open fire VOCs. In terms of spatial distribution of emissions, we

ACPD
11, C4602–C4629, 2011

can expect this contribution to follow roughly the contributions of directly emitted CO. Other work (Fisher et al., ACP, 2010; Liang et al., ACPD, 2011) takes into account CO from oxidation of co-emitted VOCs through constant scale factors. Since the goal of this paper is primarily to understand general source contributions and not perform a closed CO budget analysis, the revised paper focuses more on the fractional contributions of the direct CO emissions.

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

Our CO emissions are compared in detail with those from Fisher et al., 2010, in the revised version of the manuscript. 'Global CO sources of both anthropogenic and fi re emissions (summarized in Table 2) agree in general with the results in Fisher et al. (2010), with some differences: anthropogenic emissions for Europe are 3 Tg/month lower, US emissions are 1.5 Tg/month larger, and Asian emissions are about 3 Tg/months smaller compared to Fisher et al. (2010). Further, Russian fire emissions in April are larger (about 3 Tg/month) and Asian fi re emissions in this study are similar to Fisher et al. (2010).'

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?

We compared two simulations, one using surface emissions and one, where emissions were evenly distributed over 6km, as stated in the original version of the paper. For both simulations, emissions were effectively transported out of the boundary layer by the model, as also found in earlier studies.

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

ACPD

11, C4602–C4629, 2011

We do not agree with the reviewer. It is important to identify the range of source regions chosen in this study (new Figure 2). The text does not include this specific 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.

We agree that the presentation of CO tags in Figure 5 was misleading and changed this to relative contributions as pointed out above (see new Figure 7). We further have revised the discussion about shortcomings in the model in the new version of the paper (see comments above)

$P.5947$

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

We agree and have modified this paragraph. Other regions for errors in the model are e.g., the underestimation of local anthropogenic emissions over Alaska and Canada, as well as the underestimation of the remote influence of CO sources most likely from anthropogenic emissions (see comments above).

$P.5948$

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).

We have compared OH from MOZART-4 with the climatology from Spivakovsky et al., 2000, and see a significant underestimation of OH with a maximum in the Tropics. Since CO in mid-latitudes is well represented we assume that much the smaller error in OH at high latitudes does not impact CO at high latitudes significantly.

9. L9-10. This statement contradicts L16-17 on P. 5947, which stated " Since fire

ACPD

11, C4602–C4629, 2011

emissions are not a very important contributor for ARCTAS CO and BC measurements in early April".

See response to Reviewer 1, comments 10.

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.

We have discussed the underestimation of BC below 8 km, however, the section is rewritten and the discussion is clarified in the revised version of the paper.

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.

We agree that this statement was based on an assumption that the underestimation of anthropogenic emissions in the second and third week of April is similar to the first week in Aril. Comparisons between MOPITT and MOZART-4 at 800 hPa, do not support this statement and we have dropped it.

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

We change this number to '4-6 times'.

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.

As pointed out about, changes in the BC scavenging scheme evolve a major model

ACPD

11, C4602–C4629, 2011

development and testing exercise. Here, we discuss differences that need to be addressed in future studies.

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.

See answer to Review 1 comment 14.

$P.5950$

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.

We add this issue to the new version of the paper.

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.

This comment is not clear, at this line in the text we stated: 'high latitudes'..?

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?

We dropped Figure 7, 11, and 12. We have removed this paragraph and Figures 7,

ACPD

11, C4602–C4629, 2011

11 and 12, and instead performed a comparison between MOPITT and MOZART over source regions. Please see response to Review 1, comment iii.

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.

See comment above, the use of relative contributions is more reasonable in this context.

$P.5951$

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

See response to comment 13.

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.

See comment above, again, the use of relative contributions is more reasonable in this context.

$P.5952$

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?

The reviewer is right, but East Siberian contributions using tagged CO occur in the same altitude range as using FLEXPART trajectories (left column, second row). The model indicates a maximum in 6km, we have added this information in the revised version of the paper. 'Further, FLEXPART trajectories suggest that airmasses sampled during ARCTAS-B are strongly influenced by East Siberian fires at altitudes around 8 km in summer and from China around 10 km. This is in agreement with simulated CO tags, besides large contributions around 6 km identified using CO tags (shown in Figure 9, left column).'

ACPD

11, C4602–C4629, 2011

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.

Based on the new discussion in the revised version of the manuscript, we do not think that Canadian CO emissions are largely underestimated.

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

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.

We have removed this discussion and dropped Figures 11,12 and think they might be misleading. As discussed above, uncertainties in the model are identified and discussed more clearly in the revised version of the manuscript.

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