Response to Reviewer 2

General comments

This paper describes the modeling of CO, black carbon (BC) and SO2/SO4 in the western Arctic during the ARCTAS campaigns using the GEOS-5 model. The main goal of this modeling effort is to determine the source attributions of pollution in the Arctic using tagged CO species for different source regions and types. In general the paper is well written and provides an important contribution to pollution modeling in the Arctic, but it lacks specifics and originality and major and minor comments are listed for needed additions and improvements.

Major Comments:

1. The authors state in various places such as the introduction and conclusion that the measurements during ARCTAS are not representative for the Arctic, especially during July. Aircraft campaigns are not designed to be "representative" for specific regions by just simply averaging along the flight track. Especially during ARCTAS-B one of the major goals was to characterize boreal forest fire emissions and chemistry and therefore fire plumes were specifically targeted and clearly represent a larger fraction of the sampled air masses than the Arctic average. With a more careful analysis than just averaging along the flight track "representative" conditions can surly be found during the aircraft campaign. I suggest removing this as one major conclusion of the paper or at least add the above caveat to the manuscript.
Reply: Thanks for pointing this out. Here is the new description in the conclusion.

"The model reveals that the ARCTAS DC-8 measurements are representative of regional Arctic pollution in the spring (April, ARCTAS-A) due to relatively homogeneous tracer distribution. The aircraft data alone, however, are insufficient to provide a comprehensive and representative picture of Arctic pollution in the summer (July, ARCTAS-B) because the flights targeted local fire plumes."

2. I think the main problem with this paper is that all the source attribution is done using CO only. This has in principle been done for ARCTAS previously (Fisher et al 2010). It has been shown that for the more climate relevant and shorter-lived species BC the source attribution might be quite different from CO (Warneke et al 2010). BC has been modeled in this study as well although not as tagged species. I assume that a complete source attribution is therefore not feasible, but it might be possible to use correlations of BC with the tagged CO tracers to estimate a source attribution.
Reply: Based on the reviewer's suggestion, we analyzed the ratio of BC to CO under various source attribution environments. See new Figure 7 and the corresponding discussion in section 3.3 about model capability in characterizing source attribution.

3. A much more detailed model-measurements comparison for BC, SO2, and SO4 is also missing. A comparison is only done qualitatively for the two case studies and those two do not look very encouraging. Altitude profiles and correlation plots similar to CO should be given and main reasons for discrepancies need to be discussed with focus on the BC loss processes during the transport and the possible underestimate due to the emission inventories.
Reply: The evaluations of BC in its vertical profiles (Figure 3) have been added (also

see reply to reviewer 1 general comment 3). Sensitivity experiments for BC biomass burning emission and chemical aging have been conducted and the optimized model performances have been recommended (see section 3.1 paragraphs 4 and 5).

4. I would also like to see a more detailed description of the used emission inventories, which are given as one of the main reasons for model-measurement discrepancies. Especially the biomass burning inventory should be discussed and compared to previously used inventories for other modeling efforts such as GEOS-Chem and FLEXPART.
Reply: Table 2 was added to summarize the emissions of CO and BC from different types used by GEOS-5 for the ARCTAS campaign in section 2.3. Whenever available, GEOS-Chem CO and BC emissions and emission ranges summarized from AeroCom and hemispheric transport of atmospheric pollution (HTAP) multi-model activity were given for comparison. FLEXPART was not included for the comparison since it was used to chase plume transport. It used FLAMBE biomass burning emission, like GEOS-Chem, for CO, but used anthropogenic sources from the Center of Global and Regional Environmental Research (CGRER) only when anthropogenic emissions exceeded $1.1X10^4$ kg (CO) day^{-1} to life the background CO level [Fuelberg et al., 2010].

5. Two case studies are discussed in the paper, but I do not find the second one a good representative example for the ARCTAS campaigns. First of all both examples are from the July campaign and none from April and secondly the second case study is a transfer flight and includes lots of pollution from California, which was not the focus of the ARCTAS campaigns. I think it should be possible to find a good flight from ARCTAS-A that shows typical Arctic background air and ASFF and BOBB plumes.
Reply: We agree with the reviewer's comments and we removed the discussion for the second case study since it is a transfer flight with half of the journey outside the Western Arctic.

6. page 8835 line 18 - page 8836 line 6: For me this is the paragraph that contains the most interesting results, but it completely lacks any real discussion. It seems from Figures 3 and 4 that GEOS-5 significantly over predicts BC in clean Arctic background air. Is this because of to slow removal or over-prediction of the emission inventories? Is this also observed in April? What are the possible implications of this for climate modeling? The SO2/SO4 ratio is basically not discussed at all. The SO2 in Figure 4 does not seem to agree very well. Again, is that emission, chemistry or transport? What is also not discussed is, what this means for the modeling presented here.
Reply: The study of the second case, including the discussion in page 8835 line 18 - page 8836 line 6, has been removed (see reply above). But we added the discussion of the BC emission, aging, and removal processes for both April and July (see reply to above questions 3 and 4). GEOS-5 had a comparable removal process with GEOS-Chem and AeroCom models. The sensitivity studies for BC biomass burning emission and chemical aging, which impacts BC wet scavenging, indicate that both processes alter Western Arctic BC significantly in April. In July, local biomass burning emission becomes more important, while BC chemical aging has a minor impact on overall BC level except in clean Arctic background air where the case with a short BC aging lifetime (i.e. 1.25 days) gives a better model-observation comparison. Our optimized BC simulations (recommended in Fig. 3 and paragraph 4 Section 3.1) have improved BC values in clean Arctic background in both April and July (also see Figs. S3 and S4).

We removed the content on SO2 and SO4 since it does not add much more

information to address our objective in source attribution to the Western Arctic.

7. Various tracers, such as acetonitrile for biomass burning, were measured during ARC-TAS. How does acetonitrile compare to BOBB? How does SO2 compare to ASFF? Other halocarbons or CFCs could be used as well for Asian pollution.
Reply: Please refer to the discussion in section 3.2 and the reply to question 1 of reviewer 1 for acetonitrile for biomass burning (BB) CO and dichloromethane for fossil fuel (FF) CO. We drew the scattering plots between observed CH3CN and modeled BB CO and between observed CH2Cl2 and modeled FF CO (Fig. 4). Other halocarbons or CFCs are more useful for O3 study and the study of stratosphere and troposphere exchange. They are not used in our study of tropospheric CO and aerosols.

Minor Comments:

- Abstract: please define what is used for western Arctic.
Reply: Done.

- Chapters 3.1. and 3.2.2: These chapters seem to belong together since they both discuss the model-measurement comparison. I would re-organize the paper with: 1: comparison, 2: case studies and 3: source attribution.
Reply: The paper has been reorganized as suggested.

- page 8834 line 18ff: The first part of this flight samples what I would call Arctic background air and the authors call it clean, so I would not describe the source regions here.
Reply: This case study has been removed as suggested (see reply to the main question 5 above).

- page 8834 line 23ff: These two sentences are a repeat of what was said before and should be deleted.
Reply: This case study has been removed as suggested (see reply to the main question 5 above).

- page 8835 line 6-17: A description what BC/CO and SO2/SO4 are used for in this chapter should be moved to the introduction.
Reply: This case study has been removed as suggested (see reply to the main question 5 above).

- page 8836 line 20: What are the slopes and correlation coefficients for individual flights? The slope in Figure 5 of 0.79 is pretty encouraging, but looking at Figure 5 it seems that individual flights are clearly not as good. I think at least a range of the slopes should be given, but preferably every flight should be given.
Reply: Done. See numbers of slope and correlation for every flight in revised Figures S5 and S6.

- page 8837 line 14: While this statement seems true for total CO, is this also the case for the tagged CO species and also for BC and SO2?
Reply: We changed the subtitle to "source attribution of CO for the Western Arctic". We think the subtitle is appropriate for this work since the discussion used both total and tagged CO in section 4.

- Figures: All the Figures in the original manuscript are too small. In the supplement the Figures have about the right size.
Reply: We will make sure the figures are the right size in the published paper.

- Figure 3 and 4: The color scales should be labeled.
Reply: No long relevant.

- Figure 6: Label for the green trace is missing.
Reply: Done (now is Figure 9).