Review of "Source attribution of Arctic black carbon constrained by aircraft and surface measurements," by Xu et al.

In this paper, Xu and coauthors use the GEOS-Chem transport model to quantify the contributions from different regions to the Arctic black carbon burden during three years – 2009, 2011, and 2015. They first validate the model with surface-based monthly mean observations and with measurements from two springtime aircraft campaigns. They find relatively good agreement between the model and observed concentrations. For two Arctic sites (but not a third), this agreement improves when they include in their model an inventory of gas flaring emissions from western Siberia. Sensitivity studies with the forward model yield the contributions from different regions to Arctic BC, while simulations with the adjoint version of GEOS-Chem provide spatially-resolved information on these contributions.

The main findings of this paper are as follows: Anthropogenic BC from eastern and southern Asia dominate the Arctic BC burden in spring and contribute about one-third of the annual burden, with larger contributions aloft than near the surface. Anthropogenic BC from northern Asia are important BC in the lower troposphere, especially in spring. Biomass burning contributes 25% of Arctic BC annually. Results from the adjoint point to interesting influences on Arctic BC from regions as far south as the Indo-Gangetic Plain.

Main criticisms.

1. This paper moves forward the research on the origins of Arctic haze, providing in particular an update on how recent increases in anthropogenic BC from Asia may affect the Arctic. However, the authors do not make clear how their work builds on four recent GEOS-Chem studies that focus wholly or in part on Arctic BC: Wang et al. (2011, 2014) and Breider et al. (2014, 2017).

No doubt the authors were unaware of the 2017 paper, but the other three papers were published well before this one was submitted. Only Wang et al. (2011) is mentioned, and that only in passing. It is in particular concerning that the authors do not make clear whether they took advantage of the improvements in BC wet deposition of Wang et al. (2011, 2014). Did the authors include the snow scavenging scheme and the improvements to washout and rainout from Wang et al. (2011)? What about improvements to the impaction scavenging (Wang et al., 2014)? As is, the text cites only the wet deposition scheme of Liu et al. (2001). If the authors chose not to implement the Wang et al. (2011, 2014) improvements to wet deposition, the reader will want to know the rationale and what difference it would make if these improvements had, in fact, been included.

Brieder et al. (2014) focused on Arctic haze in 2008, and Brieder et al. (2017) examined the evolution of Arctic haze from 1980 to 2010. The authors can easily make the case that by simulating Arctic haze in 2009, 2011, and 2015, their paper provides an update to the Breider research, especially in light of increasing Asian emissions. But first they need to compare their approach and results very carefully with those in the earlier work. For example, the Brieder papers make use of a different emission inventory than does the current paper, and the reader will want to know how these inventories differ. As another example, Brieder et al. (2014)

appears to capture the mid-tropospheric peak in BC, while the current work does not. Again the reader will want to understand this discrepancy.

Responding to criticism #1 will require some effort. A close reading of the four relevant papers is necessary, and a detailed account of how the current paper moves beyond the previous papers is expected by the reader.

2. The conclusion section lacks discussion. Why should readers care about these new results? For example, what are the implications for their findings for regional climate in the Arctic? The introduction mentions some of the probable effects of BC on regional climate, and how the meteorological impacts of atmospheric BC likely differ with altitude. What does this altitude variation in forcing mean for Arctic haze of Asian origin? In addition, Brieder et al. (2017) suggests that the 1980-2010 trends in Arctic haze have contributed to regional warming. How do the new results build on Brieder et al. (2017)? How are emissions in Asia projected to change in the future, and what are the probable consequences for Arctic climate? Is gas flaring around the Arctic expected to ramp up in future decades?

3. The introduction lacks key information but is nonetheless too long. First, the authors should describe what is known about the seasonal variation of transport to the Arctic at the beginning of the paper. As is, this information appears scattered through the paper as a kind of recurring explanation for the modeled results. It would be easier for the reader to encounter this information in a succinct paragraph in the beginning, and then be reminded of how transport influences Arctic as the results emerge.

That said, the authors should condense much of the other background information in the introduction, beginning at line 14 on page 3 and continuing to the end of that section. For example, the reader doesn't need to know every published estimate of the influence of biomass burning on Arctic BC. Details of the Arctic aircraft campaigns can be saved for later in the paper.

4. The authors make much of recent increases in Asian BC emissions, but use anthropogenic emissions only for 2010 and GFED emissions for 2009, 2011, and 2014. These emissions are applied to GEOS-Chem simulations driven by 2009, 2011, and 2015 meteorological fields. The reader is curious if there are implications in using constant anthropogenic emissions and GFED emissions from a mismatched year. Also of interest is whether the authors see much interannual variation in transport over the three model years.

Minor criticisms.

Page 1, line 16. Run-on sentence.

Page 2, line 28. What is meant by "near-surface"?

Page 4, line 11. Reader is curious why published BC measurements may be biased.

Section 2.1. Years of measurements should be stated.

Page 9, line 3. The authors should consider a table providing BC emissions by region, as in Breider et al. (2014).

Page 12, line 25. Reader is confused why the measurements at Ny Alesund are halved.

Page 20, line 20. How "substantially" are shipping emissions expected to increase and over what time frame?

Page 20, line 25. The authors state: "The main difference is due to emission trends that our anthropogenic emissions from eastern and southern Asia are generally 30% higher than those in other studies." Are these increases due to increased development in Asia? Please remind the reader what time frame is being considered here.

Page 21, lines 12-21. Using the adjoint, the authors find that emissions as far south as the Indo-Gangetic Plain influence Arctic BC. This is new information. How confident are the authors of the GEOS-Chem simulation in this region (and in China)?

Figure 1. Are these total BC emissions or just anthropogenic?

Figure 3. Error bars on most measurements look very small. Please check the magnitudes. What are the years of the measurements?

Figure 4. Please put error bars on the ground-based measurements.

Figure 5. Please state in the caption the year and season of the measurements and model results.

Figure 7. Consider making a 4-panel plot with two new panels showing the stacked percent contribution of each region to the BC at different altitudes. The two new panels would have altitude on the y-axis, and percent contribution from 0-100% along the x-axis. In any case, the two existing panels look strangely elongated.

Figure 8. Measurements should have error bars.

Table 1. Table should include footnotes so that the reader does not have to scramble through the text to learn what the different scenarios mean. Also, it's not that clear that the vertical RMSE is meaningful since it varies so much with altitude.

References.

Breider, T.J., L.J. Mickley, D.J. Jacob, Q. Wang, J.A. Fisher, R.Y.-W. Chang, and B. Alexander (2014), Annual distributions and sources of Arctic aerosol components, aerosol optical depth and aerosol absorption, J. Geophys. Res. Atmos., 119, 4107-4124.

Breider, T. J., et al. (2017), Multidecadal trends in aerosol radiative forcing over the Arctic: Contribution of changes in anthropogenic aerosol to Arctic warming since 1980, J. Geophys. Res. Atmos., 122, 3573–3594.

Wang, Q., D. J. Jacob, J.A. Fisher, J. Mao, E.M. Leibensperger, C.C. Carouge, P. Le Sager, Y. Kondo, J.L. Jimenez, M.J. Cubison, and S.J. Doherty (2011), Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing, Atmos. Chem. Phys., 11, 12,453-12,473.

Wang, Q., D. J. Jacob, J. R. Spackman, A. E. Perring, J. P. Schwarz, N. Moteki, E. A. Marais, C. Ge, J. Wang, and S. R. H. Barrett (2014), Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific, J. Geophys. Res. Atmos., 119, 195–206.