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Discussions

Interactive comment on "Source attribution of Arctic black carbon constrained by aircraft and surface measurements" *by* Junwei Xu et al.

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We sincerely thank the Referee #2 for taking the time to review our paper and for providing constructive suggestions for improvement. Reponses to these comments are provided below.

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.

Response: Thank you.

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.

Response: Thanks for these suggestions. We use version 10 of GEOS-Chem, which was the latest version available at the start of this work. Thus the wet deposition of Wang et al. (2011) was implemented in our simulation. We have clarified this in text. The developments of Wang et al. (2014) were not implemented into GEOS-Chem until version 11, and thus were not included here. Furthermore, these developments have little effect in the simulations of Arctic BC as indicated by sensitivity simulations in the supporting information of Wang et al. (2014).

This manuscript is not intended to be a follow-up study of Breider et al. (2014) or Breider et al. (2017). Instead, this is an independent project (hence different emission inventories and model parameters) with different objectives. Breider et al. (2014) and Breider et al. (2017) studied major near-term climate forcers including BC in the Arctic with an emphasis on their roles in Arctic warming, whereas we aim to interpret recent measurements to investigate geographical sources and their contributions to

Arctic BC. Thus providing updates to Breider et al.'s research is not our purpose. The different emission years of Breider et al. (2014) likely contribute to differences in the middle troposphere due to different biomass burning. However, we have included a table (Table 1) with detailed regional anthropogenic and biomass burning emissions, and have given possible reasons for the discrepancy in the middle troposphere as the following to help readers understand our simulations.

"The remaining underestimation of 14 ng m-3 RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient magnitude or altitude comparisons of model with ARCTAS and ARCPAC measurements (Koch et al., 2009; Wang et al., 2011; Breider et al., 2014; Eckhardt et al., 2015) as proposed based on preferential sampling by the aircraft of plumes discussed further below."

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?

Response: Thanks for the suggestion. We have included more discussions in the conclusion as the following: "The increasing BC fraction from eastern and southern Asia at higher altitudes could have significant implications for the Arctic warming by extending the trend in increasing BC radiative forcing efficiency found by Breider et al. (2017) driven by strong increase with altitude of the direct radiative forcing of BC (Zarzycki and Bond, 2010; Samset and Myhre, 2015). Besides, anthropogenic emissions of BC in southern Asia are projected to increase under several IPCC scenarios (Streets et al., 2004; Bond et al., 2013). The climate implications of BC emissions within the Arctic are concerning given their disproportionate warming effects and the potential for increasing Arctic shipping activity as ice cover declines (Sand et al., 2013)."

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.

Response: Thanks for the suggestion. We have included the description of transport to the Arctic in the introduction as the following and have condensed the other background information.

"Analysis of observations have revealed that Arctic BC is primarily transported from regions outside the Arctic (Klonecki et al., 2003; Stohl, 2006). In winter, northern Eurasia is the primary source where air masses are cold enough to penetrate the polar dome into the Arctic lower troposphere (Stohl, 2006). Air masses from the relatively warm mid-latitudes (i.e. North America and Asia) are forced to ascend above the polar dome to the Arctic middle and upper troposphere (Law and Stohl, 2007). In spring, the warming of the surface leads to higher potential temperature over the Arctic and the northward retreat of the polar dome, facilitating the transport of air masses from mid-latitude regions to the Arctic (Stohl, 2006). However, large uncertainties remain in sources and geographical contributions to Arctic BC that require additional interpreta-

tion of observations to address."

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 inter annual variation in transport over the three model years.

Response: The time frame for the "recent" increase in Asian BC emissions is from 2000s to 2010. We have clarified this as the following in the manuscript: "The main difference is due to emission trends such that our anthropogenic BC emissions from eastern and southern Asia are generally 30 % higher than those in earlier studies (e.g. Shindell et al., 2008; Sharma et al., 2013) due to rapid development since 2000 and that our anthropogenic BC emissions in Europe are half those in prior studies due to European emission controls.".

We assume no significant change of Asian BC emissions from 2010 to 2015 because Asian BC emission growth plateaued after 2010 (Crippa et al., 2016). We also assume that using GFED 2014 emissions for 2015 simulation has little influence on our results because no abnormal forest fires have been reported for 2014 and 2015. These assumptions have been included in the manuscript on page 8 line 12-13 and page 9 line 22-23.

We do not see much inter-annual variation in transport over the three model years because the simulated vertical profiles of 2009 and 2011 campaign years are similar to each other and that the contribution from eastern and southern Asia pattern remain similar. Both 2009 and 2011 profiles show uniform (coefficient of variance of 0.08 for 2009 and 0.13 for 2011) distribution below 700 hPa and larger variation above 700 hPa. The 2015 profile exhibits a distinct enhancement in the middle troposphere that may be affected by plumes.

Minor criticisms.

Page 1, line 16. Run-on sentence.

Response: We have revised it to "Black carbon (BC) contributes to the Arctic warming, yet sources of Arctic BC and their geographic contributions remain uncertain".

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

Response: We have revised it to "Near-surface (< 1 km) BC particles".

Page 4, line 11. Reader is curious why published BC measurements may be biased. Section 2.1. Years of measurements should be stated.

Response: We have revised the sentence to "Furthermore, evidence is emerging that the BC observations to which many prior modeling studies compared may have been biased by 30 % (Sinha et al., accepted) or a factor of 2 (Sharma et al., 2017) due to other absorbing components in the atmospheric aerosol."

Years of measurements have been included in Sect. 2.1.

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

Response: Done.

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

Response: The measurements at Ny-Ålesund were not halved by us. We have clarified this as "Restricting measurements to common years changes monthly means by less than 13 %, except for a 40 % change at Ny-Ålesund in April that arises from limited data coverage in common years since PSAP measurements for April at Ny-Ålesund is not available in 2009."

Page 20, line 20. How "substantially" are shipping emissions expected to increase and

over what time frame?

Response: We have clarified these in the manuscript as "This source is expected to increase by 16 % by 2050. (Winther et al., 2014)".

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.

Response: We have clarified these in the manuscript as "The main difference is due to emission trends that our anthropogenic emissions from eastern and southern Asia are generally 30 % higher than those in earlier studies (e.g. Shindell et al., 2008; Sharma et al., 2013) due to rapid development since 2000".

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)?

Response: Emissions are a major source of uncertainty in the simulation of the contributions from the Indo-Gangetic Plain and China to the Arctic. The emissions in China and the Indo-Gangetic Plain in the HTAP v2 inventory originate from the MICS Asia inventory that represent the best estimate of emissions in Asia (Li et al., 2017). However, uncertainties still exist, so we suggested further investigations in the conclusion as the following: "The considerable impact of emissions from China and Indo-Gangetic Plain on the Arctic deserves further investigation."

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

Response: We have revised the caption to "The colormap indicates annual total BC emissions averaged over 2009, 2011 and 2015 as used in the GEOS-Chem simulation."

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

Response: Error bars of measurements at Alert were not included for the clarity of the figure, but error bars of the best estimate of BC measurements (mean EC and rBC measurements) at Alert were included. The error bar magnitudes have been corrected at Barrow and Ny Alesund. Measurement years are included in the legend.

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

Response: Done. But only one year (2009) ground measurement is available for Ny Alesund, so no error bar presents in the figure.

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

Response: Done.

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.

Response: Thanks for the suggestion. We have changed the figure to the 4-panel plot as suggested.

Figure 8. Measurements should have error bars.

Response: Done.

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.

Response: Thanks for the suggestion. We have included footnotes in Table 2. The vertical RMSE for simulations with different emissions shows the improvement with seasonal residential heating and flaring emissions in simulating vertical distributions of BC concentrations.

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