Authors' Response to Comments from the second review of Referee #2

We sincerely thank the Referee #2 for reviewing our paper for the second time and for providing constructive suggestions for further 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.

In the first revision of their paper, the authors have addressed most of the comments. The plots have improved, and the introduction and conclusion both function much better.

Response: Thank you.

Main criticism.

The authors did not respond adequately to Main Criticism #1 in my first review. That comment asked the authors to say more about how their study built on the Wang et al. (2011) and Breider et al. (2014, 2017) studies, which also investigated black carbon and its trends in the Arctic using GEOS-Chem. The authors responded:

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.

Three papers, all with the intent to validate GEOS-Chem BC in the Arctic, are not in fact "independent projects." Readers will try very hard to synthesize the results from these papers, and the authors of this paper should make that synthesis easier.

Indeed, a key goal of both Breider 2014 and Breider 2017 was to validate the GEOS-Chem simulation of Arctic BC. Validation was considered essential in the Breider papers; otherwise the radiative forcing calculated would not have been credible. Thus, readers will want to know how the new BC results differ from those of Breider and why. They will expect the current paper to compare emission inventories and model parameters with those used by Breider. Otherwise, what will the next GEOS-Chem user – or any chemistry modeler – do when she wants to model the Arctic atmosphere? What lessons can be learned? This comparison is especially crucial given the large difficulties current chemistry models have in simulating Arctic PM.

Response: Thank you for clarifying your perspective. We respectfully admit concern that undue attention is being placed on simulations from a single group, rather than on observations, on other GEOS-Chem simulations of the Arctic, or Arctic simulations by other models. Nonetheless, to address the reviewer's comments we have added additional citations of Wang et al. (2011) and Breider et al. (2014, 2017). We have carefully reread Breider et al. (2017) to follow their example in the discussion of differences from Breider et al. (2014). We now have 15 citations to these three references. Below is a list of text where Wang et al's and Breider et al's works are cited to provide evidence of how this study builds on those works.

Page 3 line 23: Some studies suggested that Europe was the dominant source of BC aloft (Stohl, 2006; Huang et al, 2010b) while others found eastern and southern Asia was the most important source (Sharma et al., 2013; Breider et al., 2014; Wang et al., 2014a; Ikeda et al., 2017) in the middle troposphere.

Page 4 line 6: BC emissions in mid- and low-latitude regions increase the Arctic climate forcing efficiency by altering the BC vertical distribution (Breider et al., 2017).

Page 5 line 3: Our work builds on knowledge gained from previous GEOS-Chem studies of Arctic BC (Wang et al., 2011; Breider et al., 2014; Breider et al., 2017; Qi et al., 2017a; Qi et al., 2017b) with major improvements including 1) new airborne measurements during 2009, 2011 and 2015 when more typical fires than in previous studies foster better understanding of anthropogenic source contributions to the Arctic; 2) new refractory BC measurements in the Arctic more accurately constrain emissions in simulations; 3) more recent and improved emissions better represent the global redistribution of BC emissions, include flaring and seasonal emissions of residential heating; and 4) seasonal source attribution using the adjoint of GEOS-Chem reveals the importance of specific sources.

Page 7 line 12: Prior Arctic aircraft campaigns (i.e. ARCTAS) were strongly influenced by the unusually extensive Russian fires in 2008 (e.g. Warneke et al., 2009; Wang et al., 2011; Breider et al., 2014). This study uses new aircraft observations when fires were less pronounced over multiple years (2009, 2011 and 2015) to better understand anthropogenic source contributions.

Page 8 line 24: The Bond et al. (2007) emission inventory for 2000 is included for comparison, since it has been widely used in modeling studies of Arctic BC (Shindell et al., 2008; Koch et al., 2009; Liu et al., 2011; Wang et al., 2011; Breider et al., 2014; Qi et al., 2017a; Qi et al., 2017b).

Page 10 line 1: Dry deposition of BC aerosols adopts a standard resistance-in-series scheme as described in Zhang et al. (2001) with improvements on BC dry deposition velocity over snow and ice following Fisher et al. (2010) and Wang et al. (2011). Wet deposition of BC aerosols is initially described in Liu et al. (2001) and developed by Wang et al. (2011) to distinguish between liquid cloud ($T > 268$ K) in which 100 % hydrophilic BC is removed and ice cloud ($T < 268$ K) in which only hydrophobic BC is removed.

Page 10 line 6: The scavenging developments of Wang et al. (2014b) are not implemented since they have little effect on Arctic BC.

Page 10 line 15: The time period simulated is 2009, 2011 and 2015, which is coincident with aircraft measurements when fires were more typical than for previous evaluations of GEOS-Chem versus

Arctic observations (i.e., Wang et al., 2011; Breider et al., 2014) to better understand anthropogenic source contributions here.

Page 14 line 11: This vertical distribution is similar to the measurements of the ARCTAS aircraft campaign in the Arctic in spring 2008 (Wang et al., 2011), though the magnitude of concentrations in this work is lower by a factor of about 2, likely because the Arctic was substantially influenced by strong biomass burning in northern Eurasia during the ARCTAS in spring 2008 (Warneke et al., 2009).

Page 21 line 13: The increasing BC fraction from eastern and southern Asia at higher altitudes could have significant implications for 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).

For example, Figures 3 and 4 of Breider 2014 reveals that adding gas flaring could indeed improve the model match with surface observations in that paper. But Breider 2014 better captures peak BC concentrations at ~5 km in spring than do any simulations in the new paper. Why is that? Is it just because of fires (Wang et al., 2011)? Or are there differences in wet deposition schemes that matter? A key conclusion of the Xu paper is that "anthropogenic emissions in eastern and southern Asia have the largest effect on the Arctic BC column burden in spring (56%)...., with the largest contribution in the middle troposphere (400-700 hPa)." If that is the case, it matters that Breider 2014 captures the BC enhancement in the mid-troposphere but the new paper does not.

Response: The mid-tropospheric peak in Breider et al. (2014) and in our work are not directly comparable due to different reasons for the peak (unusual biomass burning in Breider, possible sampling bias in this study). Furthermore, the underestimate in this study is minor (several percent). These topics are discussed further below.

One major difference in the simulation between Breider et al. (2014) and this study is fire emissions. A comparison to fire emissions in Breider et al. (2014) is confounded by the extensive tuning of that simulation to the unusual conditions of spring 2008 when boreal fires in Eurasia were unusually extensive. For example, Breider et al. (2014) scaled FLAMBE emissions from Russia by 47%, from southeast Asia by 55%, and from North America by 37.5%. The tuning used by Breider et al. (2014) is not applicable to our work since that tuning targeted the unusually extensive fire emissions in spring 2008.

Another major difference is anthropogenic emissions. Breider et al., (2014) used the Bond et al. (2007) emission inventory for 2000, with doubled emissions in Russia and Asia, while we use the HTAP emission inventory for 2010. The advantages of using the HTAP inventory over the Bond et al. (2007) inventory include 1) seasonally varying emissions of residential heating and 2) higher emissions in eastern and southern Asia that reflects the considerable growth of energy consumption in Asia over the past decades. These advantages are described in the manuscript on page 8 line 11 – 24. By using the HTAP inventory, we find that the mid-tropospheric burden is primary contributed by anthropogenic emissions in eastern and southern Asia during years (2009, 2011 and 2015) without abnormal fire activities.

Given the different reasons for the peak in the mid-troposphere (unusually extensive fires in Russia in Brieder et al. (2014); anthropogenic emissions from eastern and southern Asia in this study), we

believe these two studies are not comparable in this aspect.

Furthermore, we respectfully contend that Breider et al. (2014) did not better capture the BC peak in the middle troposphere. Figure 4 of Breider et al. (2014) showed an overestimation of BC concentrations at 4 - 6 km by 30 - 50 ng m^{-3}, while we underestimate the BC concentrations at 700 – 500 hPa by 10 – 30 ng m^{-3}.

We reiterate that the weak 'peak' in this study could be influenced by preferential sampling of plumes by the aircraft as stated on page 15.

Finally, the 'peak' raised by the reviewer is a minor issue because the difference of Arctic BC burden below 500 hPa from the simulated and the observed vertical profile is as low as 6.5%. We have added this point on page 15 line 3 as the following:
"If the measurements are representative in this region, the Arctic BC burden below 500 hPa in springtime could be 6.5 % larger than simulated here."

The authors also state:
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).

The authors should not assume that everyone knows that the developments in Wang 2014 were not implemented until v11 and in any would have little effect on Arctic BC. A key piece of writing any paper is to acknowledge what the current study lacks and then say whether or not that lack matters.

Response: Thank you. We have clarified this in the manuscript on page 10 by adding the following "The scavenging developments of Wang et al. (2014b) are not implemented since they have little effect on Arctic BC."

To describe the underestimate of the BC simulation in the mid-troposphere, the authors have added the following text:
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.

The reader is confused by "insufficient magnitude." What exactly has insufficient magntitude? The wording of the entire sentence is awkward.

Response: Thank you. We have revised the sentence to "The remaining underestimation of 14 ng m^{-3} RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient emissions or preferential sampling of plumes by the aircraft as discussed further below."

References:

Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G. and Trautmann, N. M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850-2000, Global Biogeochem. Cycles, 21(2), GB2018, doi:10.1029/2006GB002840, 2007.

Breider, T. J., Mickley, L. J., Jacob, D. J., Wang, Q., Fisher, J. A., Chang, R. Y.-W. and Alexander, B.: Annual distributions and sources of Arctic aerosol components, aerosol optical depth, and aerosol absorption, J. Geophys. Res. Atmos., 119(7), 4107–4124, doi:10.1002/2013JD020996, 2014.

Breider, T. J., Mickley, L. J., Jacob, D. J., Ge, C., Wang, J., Payer Sulprizio, M., Croft, B., Ridley, D. A., McConnell, J. R., Sharma, S., Husain, L., Dutkiewicz, V. A., Eleftheriadis, K., Skov, H. and Hopke, P. K.: 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(6), 3573–3594, doi:10.1002/2016JD025321, 2017.

Koch, D., Schulz, M., Kinne, S., Mcnaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S. and Berntsen, T.: Physics Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9001–9026, 2009.

Liu, J., Fan, S., Horowitz, L. W. and Levy, H.: Evaluation of factors controlling long-range transport of black carbon to the Arctic, J. Geophys. Res., 116(D4), D04307, doi:10.1029/2010JD015145, 2011.

Qi, L., Li, Q., Li, Y. and He, C.: Factors controlling black carbon distribution in the Arctic, Atmos. Chem. Phys., 17(2), 1037–1059, doi:10.5194/acp-17-1037-2017, 2017a.

Qi, L., Li, Q., Henze, D. K., Tseng, H.-L. and He, C.: Sources of springtime surface black carbon in the Arctic: an adjoint analysis for April 2008, Atmos. Chem. Phys., 17, 9697-9716, doi.org/10.5194/acp-17-9697-2017, 2017b.

Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, a M., Hess, P., Koch, D. M., MacKenzie, I. a., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., O.Wild, Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L. W., Jonson, J., Kaminski, J. W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J. and Zuber, a.: A multi-model assessment of pollution transport to the Arctic, Atmopsheric Chem. Phys., 8, 5353–5372, doi:10.5194/acp-8-5353-2008, 2008.

Wang, Q., Jacob, D. J., Fisher, J. A., Mao, J., Leibensperger, E. M., Carouge, C. C., Le Sager, P., Kondo, Y., Jimenez, J. L., Cubison, M. J. and Doherty, S. J.: Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing, Atmos. Chem. Phys., 11(23), 12453–12473, doi:10.5194/acp-11-12453-2011, 2011.

Wang, Q., Jacob, D. J., Spackman, J. R., Perring, A. E., Schwarz, J. P., Moteki, N., Marais, E. A., Ge, C., Wang, J. and Barrett, S. R. H.: Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific, J. Geophys. Res. Atmos., 119(1), 195–206, doi:10.1002/2013JD020824, 2014b.