

Second 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.

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.

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.

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.

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.

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 magnitude? The wording of the entire sentence is awkward.