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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Why models struggle to capture Arctic Haze: the underestimated role of gas flaring and domestic combustion emissions" *by* A. Stohl et al.

Anonymous Referee #1

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This manuscript presents a study of the sensitivity of black carbon (BC) concentrations in the Arctic to temporal resolution of emissions databases. Stohl et al. find that changes in the temporal resolution of domestic combustion emissions and the inclusion of a source from flaring from gas and oil wells substantially improve the comparison between Arctic surface measurements of BC and the model simulations. This is an interesting and useful contribution to the understanding of Arctic air pollution and the associated radiative forcing from BC both airborne and deposited to the surface. The findings suggest that further study of the emission of BC from petrochemical industry flares are needed to reduce earth system model uncertainties.

The manuscript needs revision prior to publication in ACP. The authors appear to be highly focused on the gas flaring source of BC, and may need to temper some of their conclusions. Without examining the role of the seasonality of scavenging, the authors imply that proper accounting of emissions are sufficient to explain model deficiencies in replicating the seasonal cycle at the BC at Arctic surface sites. Yet other models have been able to replicate Arctic aerosol seasonality without the enhanced emissions database used here. These differing findings need to be clearly considered in the final paper. Some changes to improve clarity also need to be made prior to publication.

Major comments:

1) The title is quite assertive–does the manuscript really explain why models (all? most? some?) "struggle" to "capture" Arctic haze? Is Arctic haze equivalent to BC concentrations, the focus of this study? I suggest a more precise title: "The underestimated role of gas flaring and domestic combustion on black carbon concentrations in the Arctic."

2) The manuscript does not discuss the recent paper by Browse et al., (Atmos. Chem. Phys., 12, 6775–6798, 2012) with the contradictory title of "The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol". Browse et al. use a global aerosol model with consideration of warm and cold scavenging processes to produce a seasonal cycle of BC and sulfate aerosol that reasonably matches Arctic surface observations, including the seasonal cycle, using only an annually varying emissions database. In their case, detailed consideration of gas flaring and monthly- or daily-varying residential sources were not necessary to get an appropriate seasonal cycle. Since the main finding of the Stohl et al. manuscript is that improved emissions incorporating gas flaring and daily-varying residential emissions are necessary to properly reproduce surface site seasonality, it would be appropriate to discuss and contrast the results with those of Browse et al. With the current manuscript, readers will be left with two competing hypotheses without a clear idea of the strengths and weaknesses of the differing approaches.

ACPD
13, C2392–C2396, 2013

3) It would be quite interesting to examine the effect of scavenging on the model results. Sensitivity tests with scavenging turned on or off would provide valuable information on the relative importance of this removal process compared with sources+transport. This would be particularly useful with respect to discussion about the Browse et al. paper.

4) Descriptions of the emissions sources are discussed in prose and in a kind of bulleted list. A more compact and clear way to present this information would be to have a table listing each emissions source, a reference, information on the temporal resolution of the database, and emission altitude. The prose could be appropriately reduced to discuss only highlights.

5) In the discussion of Fig. 8, the detailed case study of Station Nord data, the authors seem to be looking for evidence in the data and model results to support the hypothesis that natural gas flaring is an important component of the surface BC signal. For example, they state that "during this period [18-26 February], a clear direct attribution of measured EBC to flaring emissions is possible. . . ." Yet the model indicates that flaring emissions are a small fraction of the EBC in this time period. An enhancement in EBC on 24 Feb. is correlated with transport from gas flaring regions and this is pointed out as evidence for their importance. Yet a similar increase in EBC on 18-19 Feb. is NOT correlated with gas flaring sources in the model. The authors appear to be selecting time periods that support their hypothesis that gas flaring is the dominant source of BC at the Arctic surface, yet ignoring time periods that do not support this interpretation. Similarly speculative discussion is found on p. 9589: "Remarkably, the flaring contribution is largest during the first part of the episode (27-28 February), which may suggest that especially flaring emissions have been underestimated." In fact, the ratio of flaring BC to total BC in the model appears to be about constant throughout this high-EBC event. This analysis needs to be changed from a subjective evaluation of selected time periods to a quantitative analysis of the time series. For example, principal components analysis could be used to determine what fraction of the measured variance in the measured EBC is attributable to the flaring. Or multivariate regression could be

ACPD
13, C2392–C2396, 2013

similarly used. This qualitative evaluation is the main weakness of the manuscript and needs to be remedied.

6) Although likely beyond the scope of this paper, some Arctic sites, such as Barrow, have an extensive record of VOC measurements. Since oxidation of VOCs should be extremely slow in dark wintertime conditions, it might be valuable to examine these data for evidence of the very distinctive signature of oil and natural gas extraction operations. Similarly, vertical profiles of VOC measurements from the 2008 ARCTAS and ARCPAC campaigns might provide useful information on the vertical distribution of these compounds. One would expect samples taken in aged near-surface Arctic air to be enhanced in oil/gas tracer ratios when BC concentrations are elevated.

Minor comments:

A) Some of the map figures (fig. 1, 3, 6) are truly tiny–postage stamp sized. Can they be reformatted to use more space? Figure 5 is much better.

B) Shevchenko et al. is a conference abstract–not citable

C) Klimont et al. (2013) is in preparation–not citable (or accessible to the referees)

D) p. 9578 line 22. "The conversion of BC from hydrophobic to hydrophilic state. . . are (sic) ignored".)? So is the BC then always hydrophobic? If so, is it even removed by the in-cloud scavenging treatment?

E) p. 9583 line 5. Should this be "daily varying emissions" rather than "seasonally"?

F) p. 9584 line 2. Domestic emissions contribute to the *surface* BC concentrations.

G) p. 9585 line 27. Replace "bye" with "by".

H) p. 9586 line 3. Replace "confirms" with "is consistent with".

I) p. 9586 lines 8-10. Please be specific. What does "around Barrow" mean? Where did this information come from?

ACPD
13, C2392–C2396, 2013

J) p. 9586 line 23. What does "probably the most remote" mean? Is it furthest from midlatitude sources or flaring sources or biomass burning sources or . . . . Please use precise language.

K) p. 9590, line 21. Where is Vorkuta relative to source regions?

L) p. 9591 lines 10-14. This sentence is awkward and difficult to understand.

M) p. 9591 line 19. Change "other sectors than" to "sectors other than".

N) p. 9592 line 19. Two cases of possible flaring impact on surface EBC concentrations have become "several episodes" here.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9567, 2013.