

For clarity, we repeat the reviewer comments in normal font, followed by our answers in italic font.

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 residential 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.

Thank you for the overall positive assessment.

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.

The paper is indeed focused on gas flaring emissions and temporal resolution of residential combustion sources. Other models than ours are better suited to address the seasonality of wet scavenging which is also important. However, the uncertainties in the emissions, while large, are still smaller than those in the wet scavenging parameterizations. There is definitely a seasonal cycle in residential combustion emissions and this can be well described with our HDD concept, with rather small uncertainties at high latitudes. And flaring emissions, while uncertain, are a reality and have not been included in previous work. If models manage to replicate Arctic BC without seasonally varying emissions and without flaring emissions, they likely compensate these missing sources/seasonality with errors in the wet scavenging parameterizations. Our model results are far from perfect and should rather be considered as sensitivity calculations. The simulations account for seasonally varying wet scavenging due to changing precipitation and transport times with season, but use constant scavenging coefficients.

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."

BC is one important component of Arctic Haze, but there are certainly others. However, residential combustion emissions (and to some extent gas flaring) co-emit many of the other important Arctic Haze components (organic carbon, sulfate, etc.). Still, we agree that a more specific title is better and have changed it to:

“Black carbon in the Arctic: the underestimated role of gas flaring and residential combustion emissions”

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.

Thank you for the reference. It was indeed an oversight not putting our results into the perspective of this paper. We now refer to this paper in the introduction and also in the discussion section. We do not think that there are really two competing hypotheses but that both emission seasonality and scavenging seasonality are important. Our paper is important because the latter so far has received much more attention than the former. We have also added a subsection in the discussion section that specifically addresses the relative importance of emission variability vs. wet scavenging, which also includes a comparison between passive and aerosol tracers.

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.

We agree and now also present results (for brevity only for the Zeppelin research station) for simulations of our two tracers with fixed lifetimes of 3 days and 10 days and compare these to the tracer with wet scavenging.

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.

This is an interesting suggestion. However, so far there is no paper describing the emission data set we have used and we think it is necessary to go into some detail here. Furthermore, this paper focuses on the impact of emissions, so we find it appropriate to have a somewhat longer description of the emission data set used.

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

We disagree with the reviewer on this point. This is meant as a case study, not a statistical analysis. Of course, for performing case studies, we have searched for periods with influence of gas flaring at Zeppelin (not Station Nord) and in that sense this is a biased analysis. However, the purpose of the case study is more modest than what the reviewer is asking for, namely to convince the reader (and ourselves) that flaring influence is observable at an Arctic observatory. It is not meant to quantify the frequency of such events or the contribution of gas flaring to mean EBC concentrations, since we believe this is not possible with the data we have available.

However, the value of even finding a single episode with clearly attributable flaring influence is indisputable! For instance, how shall the lack of any CO/EBC correlation during the episode on 24 February be explained other than with a strong EBC source with a very low CO/EBC emission ratio? To our knowledge, there are no such sources other than flaring, even if the emission ratio for flaring is also still quite uncertain.

Our statement "during this period [18-26 February], a clear direct attribution of measured EBC to flaring emissions is possible. . . ." is indeed unfortunate, since this includes the episode on 18-19 February which is not related to flaring and which we do not discuss any further (but notice that here CO is correlated with EBC). We have changed the time period to 20-26 February, to avoid confusion.

Regarding the quantitative analysis of the time series the reviewer is asking for, we believe this is not possible (or would require data we do not have). Most importantly, flaring emissions arrive at Zeppelin almost always mixed with emissions from other sources (the 24 February episode is one of very few exceptions, and perhaps the clearest one). As soon as there is even a small contribution from other sources, it would dominate the CO signal, making a statistical analysis of CO/EBC correlations impossible. This is made even more difficult because different sources have different EBC emission ratios, so even a small EBC contribution from another source can sometimes give strong CO enhancements (and sometimes, not). Furthermore, CO has a strong seasonality, which also makes a statistical analysis difficult. However, we refer to the paper of

Hirdman et al. (2010), which identified the flaring region (though not flaring per se) as the main source region for BC in the Arctic.

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.

The reviewer is right: 1) this could be interesting; 2) it is out of the scope of this paper. It is also not entirely clear what to expect, since flaring may reduce the concentrations of many VOCs relative to when the natural gas is not flared.

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.

This is a consequence of the landscape format of ACPD. In ACP, these figures would occupy a full portrait page and thus would be much more easy to read.

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

Why should a conference abstract not be citable? Anyway, the data are now presented in full and discussed in detail.

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

Indeed, this paper is currently not yet available to the referees. There's nothing we can do to change that. However, the emission data set, including flaring layer and residential combustion layer, is accessible from the ECLIPSE (<http://eclipse.nilu.no>) and from the ECCAD portal of GEIA (<http://www.geiacenter.org>) where also a document describing key sources and features of this emission dataset is available.

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?

No, it is always assumed hydrophilic and it is removed by the in-cloud scavenging.

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

Indeed. We have changed this to daily.

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

Thanks, we have added "surface".

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

Thanks, changed.

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

Changed as suggested.

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

We meant Prudhoe Bay, where large oil activities exist and a lot of gas is flared. We point specifically to Prudhoe Bay now.

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.

Station Nord is not the northernmost station (but nearly) and it is not the farthest away from Eurasia, but probably the farthest away from all emission sources combined, as Alert is closer to North American sources. However, we have changed this sentence now to avoid using the term "most remote". For simplicity, it now just reads: "At the remote Station Nord both measured and modeled concentration levels are lower than at the other surface sites."

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

We now say: "However, we note that their sampling site was also only some 100 km from the western area associated with gas flaring, which could be an alternative explanation for the high concentrations..."

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

It has been reformulated and now reads:

"Vertical transport that is too strong or scavenging rates that are too low" and "opposite biases in these processes" in the Arctic and elsewhere have been mentioned as possible explanations for this (Bond et al., 2013). Our results suggest that the missing seasonality of residential combustion emissions as well as the lacking flaring emissions are also important.

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

Thank you. Done.

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

Well, we have investigated more than the one time period shown. But, anyway, we have replaced "several" with "individual", which is valid in any case.