Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-718-RC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temporally-Delineated Sources of Major Chemical Species in High Arctic Snow" *by* Katrina M. Macdonald et al.

Anonymous Referee #1

Received and published: 7 September 2017

This manuscript is the second to report on the results of 9-10 month long campaign (September to June) characterizing the chemical composition of fresh snow sampled at Alert. The first paper presented the data and compared it to simultaneous measurements of aerosol composition to assess the efficiency of air to snow deposition for the different analytes. Here the focus is application of PMF and the FLEXPART transport modeling tool to assess source regions for the various chemical compounds measured in the snow.

This is a solid piece of work, though I feel that the manuscript is less accessible than it could be (more on that below). I also suggest that the authors should consider changing the emphasis in several places in the discussion, to better reflect a lot of other recent (and also pioneering) work on related topics. A very good example of this arises

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as early as the abstract, where the finding that BC in the high Arctic during winter is dominantly from anthropogenic sources (fossil fuel combustion) and not biomass burning is highlighted. In section 3.2.2 their analysis refines this even more and points to sources in Eurasia for nearly all of this anthropogenic BC. To me, this is basically rediscovering some of the very early findings from a host of "Arctic Haze" investigations initiated in the 1970s which documented that the Haze was largely pollution, it was significantly absorbing due to BC, and much of it came from relatively high latitudes in Europe and Russia. Authors note that their work is focused on snow rather than aerosol, yet they explicitly assert that the snow is providing constraint on aerosol sources, so this "finding" is reassuring but perhaps not so exciting as to merit being the only factor from the PMF to be called out in the abstract. This statement about BC in the abstract notes that it is a "light-absorbing compound critical to the Arctic radiative balance" which is certainly true. However, the AMAP, 2015 assessment (cited frequently in this manuscript) points out that a suite of CTMs all agree that Asian sources dominate the atmospheric burden and climatic impact of BC in the Arctic. Most likely this apparent discrepancy is due to the highly stratified Arctic winter time troposphere, allowing Eurasian BC sources to be dominant in lower levels (sampled at surface aerosol sites and scavenged by mid- to low-level clouds) while Asian BC is at higher altitudes. In any case, I find the present result that essentially no Asian BC gets to Alert within 10 days more interesting than seeing very little biomass burning smoke in the high Arctic during winter.

A very interesting finding in this work is the lack of a strong anthropogenic sulfate signal. Arctic Haze "comprises a varying mixture of sulfate and particulate matter and, to a lesser extent, ammonium, nitrate, dust, and black carbon (e.g., Li and Barrie, 1993; Quinn et al., 2002)" (Quote from chapter 4 of AMAP, 2006; another work cited several times in this manuscript. This statement is also repeated nearly verbatim on page 2 lines 10-11 of this manuscript.) This may reflect imperfect air-snow transfer of a defining characteristic of the Arctic winter-time troposphere, greatly enhanced sulfate, or possibly strong impact from volcanic sources in this particular year (suggested by the

authors, but not very convincingly). Critically assessing air to snow transfer of sulfate would provide a nice link to the first paper in this series. However, the missing Arctic Haze sulfate signal could also reflect problems arising from sampling fresh snow from elevated snow tables (see more on this in first detailed comment below).

One final example of a finding that is perhaps misinterpreted or at least somewhat misrepresented is the attribution of PMF factor 2 to local dust. V, Se, and As are generally considered to be dominated by anthropogenic emissions, and in fact the authors point this out in their later discussion of factor 6. In particular, finding V to be enriched in Arctic Haze caused Ken Rahn to reassess, and basically refute (Rahn et al. 1985 in Atmos. Environ., see also AMAP, 2006, chapter 4), his own early suggestion that the haze was mostly dust from Asia (Rahn et al., 1977 in Nature). Mosher et al., 1993 used V to show that emissions from the generators at the DYE 3 radar station probably had a subtle but persistent impact on aerosol measurements made during the DGASP campaign. (Pretty well established that V is a tracer of oil combustion, in fact the authors point this out in discussion of factor 7.) Given the correlation between factor 2 and winds from the main station at Alert, it would seem plausible that local pollution, and not just local dust, is part of this factor.

Regarding comment about accessibility of the manuscript, the very detailed description of PMF in section 2.4.1 and section 3.1 describing how 7 factors were ultimately selected is too lengthy for a journal like ACP, especially considering that the algorithm is publicly available and presumably well described in EPA documents and Norris et al., 2014. Material in the supplemental showing the changes as additional factors are considered is well done, but not distracting to someone reading the paper who may be less interested in statistical details.

Detailed comments. The first paragraph of section 2.1 probably needs to be expanded to provide a few additional details about sampling and data screening. In particular, in Macdonald, 2017 the chemical fluxes in January and February were excluded in all analyses due to indications that the snow tables suffered extreme undercatch during

high winds in mid winter. However, in this manuscript these data are retained, the PMF is conducted on "flux per snowfall event" rather than concentration or flux per day, and spikes in several of the factors during January and February were used to support attribution of the factor to source. Authors need to justify this pretty large change in assessment of data quality (or stick with original decision and leave mid winter out of the PMF). As noted above, I wonder if low fluxes due to snow undercatch obscured the expected winter peak in sulfate flux.

Figure 3 probably needs to be modified, given its central role in attributing factors to likely sources. All 7 panels share a lot of similarities that tend to draw the eye as, or even more, strongly than small differences pointed out in the text in section 3.2. Probably the biggest problem is the bulleye very close to Alert in all of the panels. This is largely a geometric artifact reflecting that every particle released from the receptor site has to pass through a very small number of cells surrounding that site. I am pretty sure that Stohl and/or Burkhart have recognized this issue and have a recommended weighting scheme that reduces this bias (lower weights for cells closer to release site). Another minor point is that the green triangles and square in the panel for factor 7 are very hard to find (especially the Smoking Hills square). And the label under color bar should be Residence Time (not Residential), and there has to be some huge multiplier on the scale (max is not just 30 seconds)

Editorial comments by page/line number

1/23 AMAP 2011 was updated in 2017, probably should cite that report

2/6-8 Not sure how the concluding phrase about snow as a critical reservoir logically follows the first part of this sentence.

2/8-17 Given the vast literature on Arctic Haze, it is unclear how the references in this section were selected. Personally, I would like to see some of the very early work cited. At a minimum, indicate that AMAP, 2006 is a review paper and readers should see references cited therein.

3/4 The last phrase after the comma is very much a matter of personal opinion. I suggest ending sentence with a period after flux (see first detailed comment above).

3/20-21 Reword this to make argument more clear, and possibly consider different wording for "under-exaggerate". Are you saying that you tossed BDL samples to make the S/N higher than it probably should have been?

5/32 to 6/6 Is this needed? Results from PCA are not shown, and appear to be mentioned in passing just once more in the manuscript (page 8, line 8)

6/19 residential-→residence

9/1 Enhancement of Mg above the SS ratio by a factor of 1.6 is a big difference that would suggest an additional Mg source. Same is true for SO4, but excess is expected.

9/14-15 The residence time plot suggests that the middle of the GrIS is a stronger source for this factor than Norwegian Sea or North Atlantic, probably partly due to geometric artifact mentioned earlier.

10/Figure 1 Please explain what the bars on this plot are showing more clearly. What is the time component indicated by "/period" ?

12/Figure 3 Why not label the panels by source name rather than factor #?

13/29 There have been a lot of papers on emissions from fires (lab, prescribed, and wild) since 2009. Liu et al., 2017 in JGR maybe most recent. This one does not include BC, but provides access to many of the papers between 2009 and 2017. 14/1-16 Hirdman et al. 2010 (2 papers, in ACP) and Stohl et al 2006 (JGR) have shown similar. They probably should be cited.

16/1 delete "both"

16/3 delete "to"

16/3-4 There have been a lot of papers on emissions from fires (lab, prescribed, and

wild) since 2009. Liu et al., 2017 in JGR maybe most recent. This one does not include BC, but provides access to many of the papers between 2009 and 2017.

17/11 why not say “via N2O5 hydrolysis in the aerosol phase” instead of “NO3-radical chemistry”?

18/14 Laing et al. 2014 is not original source of this fact, Rahn probably closer, but maybe even he used someone else’s earlier work

18/20-21 Fact that FLEXPART rarely reaches any of these volcanoes is a little problematic.

21/7 seasonally-→seasonal

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