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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 #2

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Macdonald et al describe the results of positive matrix factorization of snow chemical composition measurement data from Alert, Nunavut in order to determine the prominent sources influencing the snow composition. Given changing Arctic source emissions with sea ice loss and increasing development, this is an important topic. A thorough description of the data analysis is provided. My main concerns, described below, surround the discussion of the results.

The main result highlighted in the abstract and conclusions is that the BC is primarily from fossil fuel burning, rather than biomass burning influence. This is not surprising since the study focuses on snow samples collected from Sept. 14, 2015 to Jun. 1, 2015, outside of the main summertime wildfire period. In several places in the paper (last paragraph of Section 3.2.3, part of Sec. 3.3, and P21 L 11-14), it is stated that

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these results “disagree” with previous snow chemical composition measurements that showed greater biomass burning influence, proving “contradicting snow BC apportionment findings”. The authors do note the influence of seasonality and changes in annual wildfire frequency and severity on contributions of biomass burning BC. However, because the references that the authors are comparing to correspond to different times and locations, a simple comparison of the percentages of biomass burning vs fossil fuel influence is not appropriate (e.g. Table 2), without an in-depth analysis of fire locations, frequency, and timing, as well as air mass trajectories associated with the various sampling sites. I would expect that the contribution of biomass burning vs fossil fuel likely depends on the site, season, and year. Therefore, I suggest revising the discussions and comparisons to provide these results as another study that points to the variability in BC source contributions, rather than suggesting that they “disagree with” or “contradict” previous results, which gives the idea of invalidating previous work, which instead may simply be different due to different timing and location. As part of this revision of the discussion, I suggest removing Table 2, or if the authors feel strongly about keeping this comparison, then information about timing, location(s), and wildfire influence (from fire maps and air mass trajectory analysis, presumably, or statements from previous papers) should be included. In addition, a more thorough literature search is needed if the authors mean for this to be a comprehensive comparison.

This is a complementary paper to the recent Macdonald et al (2017) ACP manuscript that describes the deposition of the same chemical species to the snowpack, with snow mixing ratios and fluxes of these species described. In that paper, Figure 1 shows time series over the same period of Sept 2014 to Jun 2015 for the following “key analytes” (as described in that paper), grouped according to time series correlations: Black carbon, methanesulfonate, C2O42- & NH4+, sea salt, NSS-sulfate, nitrate, NSS-K+ & NSS-Br-, and crustal metals; this is quite similar to the time series of the 7 factors (salt, dust, BC, carboxylic acids, nitrate, metals, and sulfate) in Figure 2 of the current paper. Despite this overlap, little discussion was included in the previous manuscript regarding likely sources.

The authors are encouraged to do a more thorough literature search for previous Alert snow, aerosol, and trace gas studies that likely will support their source apportionment findings and provide evidence for greater certainty for source identification. Some appropriate papers (not meant to be comprehensive) are noted below for discussion of specific factors. While not temporally resolved, Krnavek et al 2012 (Atmos. Environ.) provide a detailed source apportionment of marine, terrestrial, and atmospheric influences on Arctic surface snow composition. Most notably, the authors do not cite or compare to Toom-Sauntry and Barrie (2002, Atmos. Environ) who previously collected weekly snow samples at Alert from 1990 to 1994 and measured inorganic and organic ions; this paper is highly relevant to the current work!

Major comments:

Abstract: Currently, only two results are noted here – the names of the source factors and the fossil fuel source of the BC. Can additional results associated with other factors be mentioned here to highlight this work? Also, please be consistent between the factor names here and throughout the text (e.g. this says “regional dust”, but later it is discussed that the dust is likely local).

Section 3.1.1 Factor 1 (Marine Sea Salt): Is there seasonal dependence to the Br- enrichment factor? There is well-known multiphase bromine chemistry that occurs in the Arctic in the spring (see Simpson et al. 2007, ACP). Hara et al (2002, J. Geophys. Res.) conducted a detailed examination of Br- enrichments in Arctic aerosols and may be useful to consider for this work. A neutralization ratio of 0.8 is stated as neutral; what is the uncertainty associated with the calculated ratio? The discussion of the potential sea salt sources is muddled with respect to local vs far away sources and should be clarified, with improved flow in discussing the possibilities. Note that recent work has suggested that aerosols are not produced from frost flowers (Yang et al 2017, ACP; Roscoe et al 2011, J. Geophys. Res.). Were there are open leads upwind of the field site, such that open water was closer to the site? May et al. (2016, J. Geophys. Res.) pointed to sea salt production from leads in the fall-spring.

Section 3.2.3 Factor 3 (BC): The authors should consider the work of Doherty et al (2010, ACP), who measured light-absorbing impurities in ∼1200 snow samples across the Arctic. Dou et al (2012, ACP) previously compared measured snow BC to simulations of the spatial distribution of snow BC using the GISS-PUCCINI model. Recently, Barrett et al (2015, Environ. Sci. Technol.) used radiocarbon tracers to determine elemental carbon source apportionment between modern and fossil fuel carbon at Barrow, AK; perhaps some discussion in that work may be helpful here.

Section 3.2.4 Factor 4 (Carboxylic Acids): In the authors' consideration of carboxylic acid sources, they should consult the work of Narukawa et al (2002, Atmos. Environ.) who measured aerosol and surface snowpack dicarboxylic acids at Alert in Feb and April-May 2000. Dibb and Arsenault (2002, Atmos. Environ.) examine snow as a source of acetic and formic acids.

Section 3.2.5 Factor 5 (Nitrate): The authors cite Morin et al (2008) and Fibiger et al (2016) for nitrate cycling associated with the snowpack. However, Fibiger et al (2016) is a study at Summit, Greenland. There are other appropriate studies at Alert that should be considered in the context of the current work – for example, Ianniello et al (2002, Atmos. Environ) and Beine et al (2002, Atmos. Environ.).

Section 3.2.7 Factor 7 (Sulfate): In considering the main sources of snow sulfate, the authors should consult the work of Norman et al (1999, J. Geophys. Res.) who used sulfur isotopes to determine seasonal aerosol sulfate sources at Alert from July 1993 to Sept. 1994. The authors note that several volcanoes were active over the 2014-2015 season. This factor peaks in the early fall; does this coincide with the volcano activity and associated air mass trajectories (FLEXPART analysis)? Reorganize this section so that there is a clear flow of discussion – currently the authors go back and forth between multiple potential sources. For example, L20-21 and 27-29 seem to be somewhat contradictory as written. L6-7 on P19 seems to be tacked on and should be integrated.

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Section 3.3: This section is labeled as “Overall Apportionment”, but it is really primarily a discussion of how BC is apportioned between the factors. It may be useful to rename the title of this section, or reorganize and revise the section to make it more evenly about all of the factors. I would suggest a paragraph break at L21, with some reorganization between the two paragraphs. The authors point to mixing state of the particles potentially being important (L23-25), and this could be strengthened by citing previous Arctic studies (e.g. Weinbrunch et al 2012, Atmos. Environ.).

Table 3: It would be useful to integrate these results into the prior factor discussions (section 3.2).

Conclusions: The conclusions are very general, with limited discussion of any factor or analyte other than BC. There is an opportunity here to discuss other factors and analytes, particularly with respect to how they may change in the future, or with respect to uncertainties that should be examined in future work.

Minor Comments & Technical Corrections:

P1 L19, P8 L13, & in other locations: Please clarify text to describe the units used for calculating the percentage. I assume for BC that you are calculating the % based on mass conc? For Na+, for example, are you reporting the fraction of Na+ measured in the snow that was apportioned to the first factor? This isn’t currently clear and could be worried more clearly throughout the manuscript where percentages are used.

P1 L19: Fix phrasing/sentence structure as snow is not a light-absorbing compound.

P2 L7-10 & L14-15, P15 L15, & P17 L8-9: Provide references.

P2 L15: Please clarify the phrase “less prone to the ambiguities introduced by snowpack collection”.

P2 L27-28: Mention measurements data here – otherwise it sounds like the study includes only PMF and air mass modeling.

P7 L3: Is this supposed to be 59 samples (based on P3 L2)?

P7 L4: Clarify wording that you are discussing analyte concentrations and fluxes.

P7 L13-14: This discussion is not intuitive and could be clarified further. Can we learn about processes from these differences?

P7 L17-33, P8 L5-9, & Table 1: I suggest moving these paragraphs to the methods and supplementary information, as they discuss how the authors decided to use seven factors and do not discuss science. The section is also difficult to follow without in-depth knowledge of the method, and without referring back to the methods section frequently. Similarly, I suggest moving Table 1 to the supplementary information.

P8 L14-15: These sentences are redundant.

P9 L1-2: “Compound(s)” should be “ion(s)” here. Also, what are the uncertainties in the enrichment ratios? (These errors should be stated for all enrichment ratios reported in this manuscript.)

Figure 1: Remove “(point)” and “(bar)” on the y axes, as this is already shown in the legend, and “bar” is a unit of pressure.

Figure 1 caption: Provide further description of how to interpret the figure for improved clarity, particularly for those not familiar with PMF.

Figure 2 caption: What are the traces normalized to (themselves, other factors?)? What are the units? This caption is not clear.

P11 L9: Provide the calculated ratio in parentheses for context.

P13 L32: The neutralization equation is provided on P9, but it is not clear if the same equation is used for the calculation here and elsewhere in the paper.

Table 3: While there is a footnote defining “Southern Oceans”, I suggest renaming to Atlantic & Pacific Oceans, since “Southern Ocean” is a phrase typically referring to

near the Antarctic.

Figure 4: The abbreviation “Cbx. Ac.” In the legend is not immediately obvious; I suggested writing out “carboxylic acids” on two lines instead for improved clarity.

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