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

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Temporally-Delineated Sources of Major Chemical Species in High Arctic Snow – Response to Anonymous Referee #1

Referee comments received and published: 7 September 2017 (quoted below)

We would like to thank Referee #1 for their detailed comments and discussion. We greatly appreciate the care with which the referee has reviewed this manuscript and the improvements gained through their insight. Response to Referee Discussion Referee Comment: 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 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. Response: We thank the referee for their time in commenting on this manuscript. We agree that there is existing evidence pointing to

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a significant anthropogenic influence on particle black carbon (BC) levels in the Arctic. However, we would like to note that some recent studies specifically of the sources of BC in Arctic snow samples (i.e., Hegg et al., 2009; Hegg et al. 2010) have suggested that biomass burning is the dominant source of BC found in Arctic snow. Furthermore, as per the comment of referee #2, it has been suggested that the dominant source of BC to Arctic snow may vary by location or time of year. Thus, we think that additional evidence on the sources of BC to Arctic, specifically the portion that is deposited to Arctic snow, is important to discuss. We do agree that the manuscript would benefit by expanding the focus beyond BC. Several revisions have been made to the manuscript to give more attention to other chemical species critical to the Arctic atmosphere, as suggested above and in following referee comments. We have also added discussion on the geographic source of BC, with the findings of this paper indicating a largely central Eurasian source as opposed to an East Asian source. We would like to thank the referee for this suggestion.

Referee Comment: 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). Response: We agree with the referee that the apportionment of sulphate in this study is interesting. While the majority of sulphate is apportioned to Factor 7, sulphate, a significant mass, 24 μg/m2/period, is also apportioned to Factor 3, BC.

Compared to the mass apportionment of BC to Factor 3, 1.4 μg/m2/period, this gives a ratio of about 17 mass/mass SO42-/BC. This appears to be similar to the ratio typically observed in Arctic Haze of 10-20 mass/mass (e.g., Hopper, Worthy, Barrie, and Trivett, 1994; Sharma, Lavoué, Chachier, Barrie, and Gong, 2004; Gong et al., 2010 to name a few). Thus, the SO42- apportioned to Factor 3, BC, seems appropriate for Arctic Haze. Furthermore, SO42- was observed to have significant mass loading of 46 μg/m2/period on Factor 6, non-crustal metals, also considered to be anthropogenic in origin. As the referee noted, the previous publication Macdonald et al. (2017) found SO42- to show a higher deposition velocity than BC, especially in the warmer fall months. Several factors likely contributed to this trend. A potential explanation could be that heightened SO2 scavenging in the fall lead to an increased level in the snow relative to BC. Specifically sulfate/SO2 from volcanic sources prevalent in the fall may have been scavenged more readily than BC, resulting in an enhanced SO42- deposition velocity and the identification of a separate SO42- dominated factor in the fall. Additional research would be required to confirm this hypothesis (i.e. the SO2 would have to be oxidized to sulfate in the precipitation or snow), but we believe it is a reasonable explanation of the observations of these two papers. The discussion of Factor 7 has been revised to expand on these points and we would like to thank the referee for their suggestion. Please see the response to the first detailed comment for a discussion on the impact of undercatch on the sulphate signal. Gong, S. L., Zhao, T. L., Sharma, S., Toom-Sauntry, D., Lavoué, D., Zhang, X. B., Leaitch, W. R., and Barrie, L. A.: Identification of trends and interannual variability of sulfate and black carbon in the Canadian High Arctic: 1981-2007, J. Geophys. Res.-Atmos., 115 (D07305), 1–9, doi:10.1029/2009JD012943, 2010. Hopper, J. F., Worthy, D. E. J., Barrie, L. A., and Trivett, N. B. A.: Atmospheric observations of aerosol black carbon, carbon dioxide and methane in the high arctic, Atmos. Environ., 28, 3047–3054, doi:10.1016/1352-2310(94)90349-2, 1994. Sharma, S., Lavoué, D., Chachier, H., Barrie, L. A., and Gong, S. L.: Long-term trends of the black carbon concentrations in the Canadian Arctic, J. Geophys. Res.-Atmos., 109 (D15203), 1–10, doi:10.1029/2003JD004331, 2004.

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Referee Comment: 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. Response: While we agree that V, Se, and As are typical of anthropogenic sources they also occur in dust sources. The ratio of these metals to Al in Factor 2, crustal metals, were 0.0016, 0.0031, and 0.00081 m/m for V, Se, and As, respectively. Soils vary significantly in composition, but typical ratios to Al are 0.0012 - 0.0016, 0.000001 - 0.00027, and 0.00002 m/m for V, Se, and As, respectively (Taylor, 1964; Barrie, den Hartog, and Bottenheim, 1989; Masson-Delmotte et al., 2013). Measurements of local crustal sources in the Arctic have also seen ratios to Al of 0.0013 and 0.00013 m/m for V and As, respectively (Se not measured) (Barrie, den Hartog, and Bottenheim, 1989). As discussed in the manuscript, this gives enhancement ratios of approximately unity for V, 11-5000 for Se (note this large range is a result of the high variability in crustal measurements), and 6-37 for As. Thus, the loading of V in particular on this factor is very reasonable for a crustal source. The loadings of Se and As are higher than for typical soils but given the variability seen across crustal sources both could still be explained by a crustal source. Furthermore, the raw unapportioned concentration measurements of V, Se, and As all correlate to Al with Pearson's correlation coefficients of 0.91 or higher. Timeseries of these analytes are provided in the supplemental. An important distinction in this analysis is that the

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V, Se, and As measurements being discussed are the insoluble portions (as noted in the original manuscript page 7 lines 31-32, and revised manuscript page 7 lines 2-3). The soluble portion of these metals was often below detection limits with weak signal-to-noise and therefore was excluded from the apportionment analysis (note that the portion considered as "soluble" would include soluble metals as well as insoluble metals associated with particles capable of passing through a 0.45 μm filter; Macdonald et al., 2017 provides further details about this analysis). Of these three metals soluble As had the highest number of measurements about detection limit. The soluble As time series correlated best with Factors 3 and 6, black carbon and non-crustal metals. The limited data available for soluble metals contributes a high degree of uncertainty to any discussion of their potential apportionment, but their correlation with these anthropogenic factors may indicate that the anthropogenic sources of these metals were mostly captured in the soluble measurements while the insoluble measurements represent a largely crustal source.

Referee Comment: 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. Response: We agree with the referee that a detailed description of PMF is not required within the manuscript, given the target audience of this paper. Though we do think that this information is vital to be included in all papers with PMF analyses for reproducibility and transparency. Thus, portions of section 2.4.1 and 3.1 have been moved to the supplemental. âĂČ Response to Detailed Comments Referenced to Page/Line #(s) in the original manuscript: 2/31-3/4 Referee Comment: 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. Response: Additional details on the sampling procedure have been provided in the supplemental, revised section S1. We do not believe the undercatch noted in the previous study detrimentally impacted this sourcing analysis. The composition of the snow throughout January and February is not expected to be impacted by undercatch, simply the total volume of snow. Underestimation of all analytes for a few dates does not greatly impact the apportionment of a PMF analysis, since this analysis focusses primarily on the relative variation in analytes rather than their magnitude. The profiles of the identified factors should be largely unaffected; however, the temporal flux contributions may be underestimated across all factors for the dates of interest. Furthermore, the source regions identified for each factor by weighted FLEXPART analysis may have understated the impact of source regions prevalent on those dates, but the peaks identified outside of this period should not be affected and are still valid episodes. To better understand the impact of using snow flux instead of concentration three PMF analyses were completed: based on snow concentration, flux per period, and flux per day. The results of these auxiliary runs have been provided in this paper, moved to the supplemental per the referees’ suggestions. The concentration PMF factor profiles were found to be highly consistent with those of the flux per snowfall analysis considered in the manuscript. The factor compositions agreed with Pearson’s correlation coefficients of 0.97 or higher and contributions agreed with correlations of 0.60 or higher. If the uncertain January and February dates were removed the correlation of the factor contributions between the concentration and flux per snowfall

PMF analyses only changed by less than 6%. Furthermore, the primary evidence used in the identification of the PMF factors in this manuscript was composition, which does not appear to have been impacted by the underestimation of flux based on undercatch in January and February. Specifically looking at sulphate, the concentration time series is very similar to that presented for flux, with a Pearson's correlation of 0.76. Both show a very distinct fall peak with small episodic peaks in winter and spring. Neither show the typical Arctic Haze trend with a broad peak throughout the winter, as observed for BC. For reference, the concentration PMF results are provided in the supplemental and a complete record of the measured concentrations provided in Macdonald et al. (2017). Overall, we chose to include these time periods so as to not lose potential information about sources during this important time of the year. A brief note on this topic has been added to the revised manuscript. (revised manuscript page/line(s): 3/15-17) Figure 3 Referee Comment: 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) Response: While we agree that the plots in Figure 3 do share some similarities, we do not believe this is reason to change them. We agree that the "bullseye" on Alert is the result of all tracers being initialized at this location; however, it is correct to say that Alert and the surrounding area is a significant potential source/influencing area for all factors. Reducing the weighting on this area may help in identifying long-range sources but we believe it is important to emphasize that all factors could potentially be strongly influenced by local activities. Also, we find it interesting that some factors seem to show common source/influence areas. Specifically, Factors 3, 5, and 6 all likely have anthropogenic origins and all show similar source regions, with some small exceptions. These source regions show a distinct contrast from those of Factors 1, 2, 4, and 7 which appear to be more dependent on Arctic sources/influences. We agree that the symbols denoting Alert and volcanic sources are quite small (as noted by both referee #1 and 3). This was done so as to not block a significant portion of the trajectory plot. This figure will be uploaded as a high-resolution image allowing readers with difficulty seeing these symbols to simply zoom in as needed, without sacrificing the details of the trajectory plot. The legend has been corrected to residence time. The scale has been converted to a unitless relative residence time since interpretation of the actual residence time requires information on the cell size. (revised page 10) 1/23 Referee Comment: AMAP 2011 was updated in 2017, probably should cite that report Response: We thank the referee for this note. The reference to AMAP 2011 has been updated to the 2017 revision and this revision reviewed for any changes in relevant sections. 2/6-8 Referee Comment: Not sure how the concluding phrase about snow as a critical reservoir logically follows the first part of this sentence. Original Line: Particles entering the Arctic atmosphere can be removed only by atmospheric transport or deposition, and the deposition processes are much slower in the winter than in the summer; thus Arctic snow is a critical reservoir within the Arctic system. Response: We agree that this line was poorly phrased. The line has been revised to clarify as follows: Particles entering the Arctic atmosphere in winter can be removed only by atmospheric transport or deposition in snow where they can be retained for an extended time; thus Arctic snow is a potentially critical reservoir within the Arctic system. (2/17-19) 2/8-17 Referee Comment: 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. Response: We

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agree that additional sources should be included, but recognize that this is not meant to be comprehensive review paper. The following references have been added to text; furthermore, we have urged the reader to see the references within existing review papers for further information. Barrie, L. A.: Arctic air pollution: An overview of current knowledge, Atmos. Environ., 20 (4), 643–663, doi:10.1016/0004-6981(86)90180-0, 1986. Mitchell, J. M.: Visual range in the polar regions with particular reference to the Alaskan Arctic, J. Atmos. Terr. Phys., 17, 195–211, 1957. Rahn, K.A., Borys, R., and Shaw, G. E.: The Asian source of Arctic Haze bands, Nature, 268, 713–715, doi:10.1038/268713a0, 1977. Shaw, G., and Wendler, G.: Atmospheric turbidity measurements at McCall Glacier in northern Alaska, B. Am. Meteorol. Soc., 53 (5), 510, 1972. 3/4 Referee Comment: 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). Original Line: The use of a snow table allowed the deposition area associated with each sample to be recorded and used in the conversion of measured concentration to flux, which provided a considerable advantage over previous snow sampling campaigns. Response: This line has been revised per the referee's suggestion. (3/13-15) 3/20-21 Referee Comment: 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? Original Line: The signal-to-noise (S/N) of each analyte was also calculated to indicate the strength of each measurement. Given the enhanced uncertainty of below MDL and missing values, these data points were excluded so as to not under-exaggerate the S/N (Norris et al., 2014). Response: The calculation for signal-to-noise was adopted from the EPA PMF guide (Norris et al., 2014 equation 5-3 and 5-4) and is suggested for environmental data. This approach is meant to recognize that environmental data often include some missing or even negative values which, with the older PMF4 S/N calculation, would have artificially decreased the S/N ratio. This line was revised to clarify. (4/3-6) 5/32-6/6 Referee Comment: 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) Response: The paragraph mentioned provides a description of the principal component analysis and how it was applied to this data. As the referee notes, the results of this analysis are only provided in the supplemental and are only briefly discussed in the text. Per the referee's suggestion the bulk of this paragraph has been moved to the supplemental, section S4.3. 6/19 Referee Comment: residential-→residence Response: Editorial comment addressed in revised text. (6/3) 9/1 Referee Comment: 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. Response: The enrichment of Mg2+ and SO42- has been noted in the text. The enrichment of Mg2+ was found to be consistent even for PMF analyses with a greater number of factors which does not suggest a missing factor is responsible for the enrichment. Furthermore, similar enrichment of Mg2+ in a sea salt factor was also observed by Krnavek et al. (2012). The uncertainty of these enrichment ratios has been included in the text, presented as the PMF 25th and 7th bootstrapping results. (11/11-18) Krnavek, L., Simpson, W. R., Carlson, D., Domine, F., Douglas, T. A., and Sturm, M.: The chemical composition of surface snow in the Arctic: Examining marine, terrestrial, and atmospheric influences, Atmos. Environ., 50, 349–359, doi:10.1016/j.atmosenv.2011.11.033, 2012. 9/14-15 Referee Comment: 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. Response: It has been noted in the text that the influence of the area immediately around Alert may be over-exaggerated in Figure 3. While it is true that the Greenland ice sheet is a potential area of influence for Factor 1, the ice-free Norwegian sea and Northern Atlantic ocean are also potential areas of influence and we believe are a more probable potential source region. 10/Figure 1 Referee Comment: Please explain what the bars on this plot are showing more clearly. What is the time component indicated by "/period"? Original Line: Factor profiles. Error bars show the 25th and 75th percentiles of the bootstrapping analysis. Flux contributions below 0.00001 μg/m2/period are not shown. Response: The percentile and mass loading to each factor is the typical method of describing PMF results. A thorough discussion of how to interpret these results is provided in the EPA PMF guide (Norris et al., 2014). However, the author recognizes that not all readers will be familiar with such analyses. For clarity, the Figure 1 caption has been revised as follows: Factor profiles. The loading of each analyte to each factor is provided as the portion of their flux apportioned to that factor as well as the percentage of the analyte's total flux (mass/mass) apportioned to that factor. Error bars on the percentage loading show the 25th and 75th percentiles of the bootstrapping analysis. Flux contributions below 0.00001 μg/m2/period are not shown. Metals with a charge are those measured by IC, others are insoluble portions measured by ICP-MS. (revised page 8) Section 3.1 paragraph one describes the flux per snowfall period metric used. 12/Figure 3 Referee Comment: Why not label the panels by source name rather than factor #? Response: Figure 3 has been updated to include full factor names. (revised page 10) 13/29 Referee Comment: 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. Response: This section has been revised to include references to the following more recent studies: (13/31-32) Liu, X., Huey, L. G., Yokelson, R. J., Selimovic, V., Simpson, I. J., Müller, M., Jimenez, J. L., et a;.: Airborne measurements of western U.S. wildfire emissions: Comparison with prescribed burning and air quality implications, J. Geophys. Res. Atmos., 122, 6108–6129, doi:10.1002/2016JD026315, 2017. May, A. A., McMeeking, G. R., Lee. T., Taylor, J. W., Craven, J. S., Burling, I., Sullivan, A. P., et al.: Aerosol emissions from prescribed ïňĄres in the United States:A synthesis of laboratory and aircraftmeasurements, J. Geophys. Res. Atmos.,119,11,826–11,849, doi:10.1002/2014JD021848, 2014. 14/1-16 Referee Comment: Hirdman et al. 2010 (2 papers, in ACP) and Stohl et al 2006 (JGR) have shown similar. They probably should be cited. Response: The following references have been added to Section 3.2.3: (15/3) Hirdman, D., Burkhart, J. F., Sodemann, H., Eckhardt, S., Jefferson, A., Quinn, P. K., Sharma, S., Ström, J., and Stohl, A.: Long-term trends of black carbon and sulphate aerosol in the Arctic: Changes in atmospheric transport and source region emissions, Atmos. Chem. Phys., 10, 9351–9368, doi:10.5194/acp-10-9351-2010, 2010. Stohl, A., Berg, T., Burkhart, J. F., Fjæraa, A. M., Forster, C., Herber, A., Hov, Ø., et al.: Arctic smoke – record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe, Atmos. Chem. Phys., 7, 511–534, doi.org/10.5194/acp-7-511-2007, 2007. 16/1 Referee Comment: delete “both” Response: Editorial comment addressed in revised text. (15/20) 16/3 Referee Comment: delete “to” Response: Editorial comment addressed in revised text. (15/22) 16/3-4 Referee Comment: 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. Response: See response to detailed comment 13/29 above. 17/11 Referee Comment: Why not say “via N2O5 hydrolysis in the aerosol phase” instead of “NO3-radical chemistry”? Original Line: The mid-winter peak in this factor may be linked to NO3- formation via NO3-radical chemistry, which is considered to dominate Arctic NO3- chemistry during the night (Morin et al., 2008). Response: This line was revised as suggested. (17/1-2) 18/14 Referee Comment: Laing et al. 2014 is not original source of this fact, Rahn probably closer, but maybe even he used someone else’s earlier work Original Line: Non-crustal Se is typically considered to be a tracer of coal combustion and V a tracer of oil combustion (Laing et al., 2014). Response: We agree that the original reference should be provided. The following references have been added, which we believe to be some of the earliest to discuss this topic. (18/11-12, 20/1-2) Key, C. W., and Hoggan, G. D.: Determination of trace elements in fuel oils, Anal. Chem., 25 (11), 1673–1676, doi:10.1021/ac60083a027, 1953. Rahn, K. A.: Sources of trace elements in aerosols – An Approach to clean air, Ph.D. thesis, University of Michigan, 1971. 18/20-21 Referee Comment: Fact that FLEXPART rarely reaches any of these volcanoes is a little problematic. Response: The Factor 7, Sulphate, section has been revised to address several comments from all referees. We recognize that Figure 3 does not show high influence from the noted volcanic sources for Factor 7; however, this plot only represents a ten-day back trajectory and does seem to indicate that Factor 7 is more likely a dominated by relatively local sources rather than long-range anthropogenic sources. Furthermore, these plots only highlight areas over which the trajectories passed within 500 m of the surface (as noted in section 2.4.2). This approach is useful for identifying ground-level sources which could have reasonably impacted the air mass. However, volcanic sources can impact air masses to a much great height, given the heat and velocity of the emitted plume; thus, trajectories at a greater height should be considered. We have reviewed the FLEXPART influence plot for Factor 7 for trajectories within 10 km of the surface and this plot does show greater potential influence from the BárÃřarbunga volcano in Iceland and the Smoking Hills in Canada. (section 3.2.7) 21/7 Referee Comment: seasonally-→seasonal Response: Editorial comment addressed in revised text. (22/22)

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-718/acp-2017-718-AC1-supplement.pdf

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