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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 #2

Referee comments received and published: 25 September 2017 (quoted below)

We would like to thank Referee #2 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: 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 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. Response: We agree with the referee that the discussion and tone of the listed sections should be changed. While we did try to keep our literature comparison to mostly studies of similar seasons and locations, we do agree that the sources of

BC appear to be dependent on several factors. The paper has been revised to avoid statements that these results contradict those of previous studies and we instead state that they highlight the importance of understanding the variability of BC sources to Arctic snow. In general, the focus of the paper has been shifted away from BC, per the comments of the referees. Furthermore, Table 2 of the refereed document has been removed. This table was meant to be illustrative rather than comprehensive, but we agree that it is not needed.

Referee Comment: 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. Response: This manuscript is meant to be a companion to the previous paper (Macdonald et al., 2017) mentioned by the referee. The first paper outlines the measurements and analysis in greater detail and provides a comparison with concurrent atmospheric measurements. This paper expands on the previous, focussing on sources of these analytes to Arctic snow. Per the suggestion of the referees some additional references to the first paper and over-arching discussion have been added to the revised manuscript. The time series provided in Macdonald et al. (2017) are grouped into related species or those with similar measured ranges, to facilitate plotting. All apportioned time series are also provided in this paper’s supplemental. Macdonald, K. M., Sharma, S., Toom, D., Chivulescu, A., Hanna, S., Bertram, A. K., Platt, A., Elsasser, M., Huang, L., Tarasick, D., Chellman, N., McConnel, J., Bozem, H., Kunkel, D., Ying Duan, L., Evans, G. J., and Abbatt, J. P. D.: Observations of atmospheric chemical deposition to high Arctic snow, Atmos.

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Referee Comment: 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! Response: The study by Toom-Sauntry and Barrie (2002) is referenced in the previous paper discussing these snow measurements (Macdonald et al., 2017). A comparison of the snow measurements from this campaign to those in previous studies, including Toom-Sauntry and Barrie 2002, is included in Macdonald et al. (2017) supplemental section S1. The trends and absolute values of major ions measured in snow in this study were mostly found to be consistent with those observed by Toom-Sauntry and Barrie, 2002. However, we agree that further discussion of how these measurements compare to those of Toom-Sauntry and Barrie within this paper is also warranted. Sections 3.2.1 and 3.2.7 have been revised to include this discussion. We thank the referee for suggesting Krnavek et al. (2012). We have reviewed this paper and incorporated it into our discussion. We have also expanded our literature review of other related studies. The following references have been added to the manuscript: Barrett, T. E., Robinson, E. M. Usenko, S. and Sheesley, R. J.: Source contributions to wintertime elemental and organic carbon in the western Arctic based on radiocarbon and tracer apportionment, Environ. Sci. Technol., 49 (19), 11,631–11,639, doi:10.1021/acs.est.5b03081, 2015. Breider, T. J., Mickley, L. J., Jacob, D. J., Wang, Q., Fisher, J. A., Chang, R. Y.-W., and Alexander, B.: Annual distributions and sources of Arctic aerosol components, aerosol optical depth, and aerosol

absorption, J. Geophys. Res.-Atmos., 119, 4107–4124, doi:10.1002/2013JD020996, 2014. Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, a. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, Atmos. Chem. Phys., 10, 11,647–11,680, doi:10.5194/acp-10-11647-2010, 2010. Dou, T., Xiao, C., Shindell, D. T., Liu, J., Eleftheriadis, K., Ming, J., and Qin, D.: The distribution of snow black carbon observed in the Arctic and compared to the GISS-PUCCINI model, Atmos. Chem. Phys., 12, 7,995–8,007, doi:10.5194/acp-12-7995-2012, 2012. 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. 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. Law, K. S., Stohl, A., Quinn, P. K., Brock, C. A., Burkhart, J. F., Paris, J.-D., Ancellet, G., et al.: Arctic air pollution: New insights from POLARCAT-IPY, B. Am. Meteorol. Soc., 95 (1), 1873 – 1895, doi:10.1007/BF00138862, 2014. McConnell, J. R., Edwards, R., Kok, G. L., Flanner, M. G., Zender, C. S., Saltzman, E. S., Banta, J. R., et al.: 20th-Century industrial black carbon emissions altered Arctic climate forcing, Science, 317, 1381–1384, doi:10.1126/science.1144856, 2007. Pratt, K. A., Custard, K. D., Shepson, P. B., Douglas, T. A., Pöhler, D., General, S., Zielcke, J., et al.: Photochemical production of molecular bromine in Arctic surface snowpacks, Nat. Geosci., 6 (5), 351–356, doi:10.1038/ngeo1779, 2013. Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and Maksyutov, S.: 16-year simulation of arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition, J. Geophys. Res.-Atmos., 118, 943–964, doi:10.1029/2012JD017774, 2013. 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.

Toom-Sauntry, D. and Barrie, L. A.: Chemical composition of snowfall in the high Arctic: 1990–1994, Atmos. Environ., 36, 2683–2693, doi:10.1016/S1352-2310(02)00115-2, 2002. VanCuren, R. A., Cahill, T., Burkhart, J., Barnes, D., Zhao, Y., Perry, K., Cliff, S., and McConnell, J. R.: Aerosols and their sources at Summit Greenland - First results of continuous size- and time-resolved sampling, Atmos. Environ., 52, 82–97, doi:10.1016/j.atmosenv.2011.10.047, 2012 âĂČ Response to Detailed Comments – Major Comments Referenced to Page/Line #(s) in the original manuscript: The first paragraph Abstract Referee Comment: 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). Response: Per the referees’ suggestion, the abstract has been revised to briefly summarize all factors resolved rather than focussing on Factor 3, BC. We agree that factor naming should be consistent throughout. The revised manuscript uses the following names when referring to Factors 1 to 7, respectively: sea salt, crustal metals, black carbon, carboxylic acids, nitrate, non-crustal metals, and sulphate. Section 3.2.1 – Factor 1 (Marine Sea Salt) Referee Comment: 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. Response: Br-enrichment is observed in the spring. This observation was discussed in the previous companion paper: Macdonald et al., 2017. The time series of Br- is provided in the supplemental, showing a broad spring peak, and mentioned in the manuscript in section 3.2.5. This peak is not well-predicted by the PMF results. Section 3.2.1 of the revised manuscript has been updated to include a brief mention of Br-enrichment. The neutralization ratio of each factor is summarized in the revised Table 2. This table also includes the ratio calculated from the 25th and 75th bootstrapping. Factor 1 has a neutralization ratio of 0.79 with bootstrapping of 0.75 to 0.84. Section 3.2.1 discussion has been revised to improve flow and clarity. The correlation between Factor 1, sea salt, and local wind speeds was weak, a Pearson's correlation of 0.28. We agree that for local wind speeds to be relevant there must be a local source of sea salt. This could include any local open water, blowing saline snow, or frost flowers; however, we would require more data to confirm the existence of any of these sources at the specified time. The possibility of a frost flower source has been noted as quite uncertain in the revised text. Upon further consideration, we have noted that Factor 1, sea salt, in fact has a stronger correlation with collection period length (Pearson's correlation coefficient of 0.47). The January peak of this factor was one of the longer collection period of the campaign. This may suggest that the deposition of sea salt aerosol was relatively continuous over time; thus, longer collection periods were associated with higher sea salt signatures. However, it should be noted that both of these correlations are fairly weak, so these inferences should be considered uncertain. The 0.28 correlation between Factor 1 and wind speeds has been deemed too weak to include in the revised manuscript (a minimum of 0.3 has been imposed on the values included). Section 3.2.3 – Factor 3 (BC) Referee Comment: The authors should consider the work of Doherty et al (2010, ACP), who measured light-absorbing impurities in âĹij1200 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. Response: The suggested references have been added to section 3.2.3. Section 3.2.4 – Factor 4 (Carboxylic Acids) Referee Comment: 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. Response: Narukawa, Kawamura, and Bottenheim (2002) explored dicarboxylic acid measurements in Arctic aerosol and surface snowpack. Given that this campaign did not include measurements of formate and/or acetate which are the dominant components of Factor 4, we have decided not to include it in the discussion. However, we thank the referee for his suggestion. The Dibb and Arsenault (2002) paper mentioned is already included in this discussion. (16/1-2) Dibb, J. E. and Arsenault, M.: Shouldn't snowpacks be sources of monocarboxylic acids?, Atmos. Environ., 36, 2513–2522, doi:10.1016/S1352-2310(02)00131-0, 2002. Section 3.2.5 – Factor 5 (Nitrate) Referee Comment: 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.). Response: The suggested references have been added to the manuscript: Beine, H. J., Honrath, R. E., Domine, F., and Simpson, W. R.: NOx during background and ozone depletion periods at Alert: Fluxes above the snow surface, J. Geophys. Res., 107 (D21), 7-1–7-12, doi:10.1029/2002JD002082, 2002. Ianniello, A., Beine, H. J., Sparapani, R., Di Bari, F., Allegrini, I., and Fuentes, J. D.: Denuder measurements of gas and aerosol species above Arctic snow surfaces at Alert 2000, Atmos. Environ., 36 (34), 5,299–5,309, doi:10.1016/S1352-2310(02)00646-5, 2002. Section 3.2.7 – Factor 7 (Sulfate) Referee Comment: 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. Response: Sirois and Barrie (1999), the companion paper to Norman et al. (1999) provides further analysis of aerosol sources. This study is cited within the manuscript. Section 3.2.7 has been revised to improve flow and clarity. The text does state that BárÃřarbunga, a volcano in Iceland, was active during the observed fall peak. The revised section gives details on how this compares with the FLEXPART analysis. Lines 20-21 and 27-29 of the original manuscript have been removed in the revision. Section 3.3 Referee Comment: 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.). Response: Section 3.3 has been heavily revised to include greater discussion of all factors and reduce focus on Factor 3, BC. The apportionment of all analytes has been summarized in the revised Table 3. Figure 4 has also been expanded to show the apportionment of BC, SO42-, and insoluble V. Table 3 Referee Comment: It would be useful to integrate these results into the prior factor discussions (section 3.2). Response: Per the referee’s suggestion Table 3 has been moved to revised section 3.2. (now Table 2). Conclusions Referee Comment: 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. Response: The conclusions have been revised to discuss other factors and analytes in greater detail. âĂČ Response to Detailed Comments – Minor Comments and Technical Corrections Referenced to Page/Line #(s) in the original manuscript: 1/19, 8/13, and other locations Referee Comment: 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. Response: The referee is referring to the percentile loadings of various analytes onto each PMF factor. This represents the portion of total analyte mass apportioned to a single factor. To clarify, “mass/mass” has been added where appropriate, and the first use has been described as follows: The first factor was characterized by high loadings (>75% of total flux mass apportioned to Factor 1) of Na+ and Cl- and 30-45% loadings of Br-, K+, and Mg2+ (Figure 1; Table 2). (revised manuscript page/line(s): 11/8-10) 1/19 Referee Comment: Fix phrasing/sentence structure as snow is not a light-absorbing compound. Original Line: The majority (73%) of the black carbon in snow, a light-absorbing compound critical to the Arctic radiative balance, was found to be the product of fossil fuel burning with limited biomass burning influence. Response: Per the comment above, the abstract has been revised to provide further details about all factors, with less focus on BC. The line above has been removed from the revised abstract. 2/7-10 & 14-15, 15/15, and 17/8-9 Referee Comment: Provide references. Original Lines: 2/7-10: 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. Given the seasonal variability in Arctic aerosol inputs and outputs, a period of enhanced accumulation is typically experienced during the Arctic winter and early spring termed “Arctic Haze”. 2/14-15: However, direct measurements of pollutants in Arctic snow have been less common, particularly sampling campaigns of fresh snow which are less prone to the ambiguities introduced by snowpack collection. 15/15: Possible contributors hypothesized in other studies of arctic carboxylic acids are discussed below including biomass burning, atmospheric or snow photochemical processing, and ocean microlayer emissions. 17/8-9: February to June, 2015, was also characterized by a "bromide explosion", observed as a broad peak in snow and atmospheric Br-. Response: The following reference has been noted to the revised manuscript for Line 2/7-10 (revised page/line: 2/19) AMAP: Acidifying pollutants, Arctic haze, and acidification in the Arctic, Arctic Monitoring and Assessment Programme, Oslo, Norway, 2006. Line 2/14-15 has been revised as follows: "However, direct measurements of pollutants in Arctic snow have been less common, particularly sampling campaigns of fresh snow." per the comment below. (2/26-27) Line 15/15 is simply listing topics that will be discussed in the following section. The following references are provided in the following discussion: Jaffrezo, J.-L., Davidson, C. I., Kuhns, H. D., Bergin, M. H., Hillamo, R., Maenhaut, W., Kahl, J. W., and Harris, J. M.: Biomass burning signatures in the atmosphere of central Greenland, J. Geophys. Res., 103, (D23), 31067-3108, doi:10.1029/98JD02241, 1998. Legrand, M., and de Angelis, M.: Origins and variations of light carboxylic acids in polar precipitation, J. Geophys. Res., 100 (Di), 1445–1462, doi:10.1029/94jd02614, 1995. The following reference has been noted to the revised manuscript for Line 17/8-9 (16/32-33) Macdonald, K. M., Sharma, S., Toom, D., Chivulescu, A., Hanna, S., Bertram, A. K., Platt, A., Elsasser, M., Huang, L., Tarasick, D., Chellman, N., McConnel, J., Bozem, H., Kunkel, D., Ying Duan, L., Evans, G. J., and Abbatt, J. P. D.: Observations of atmospheric chemical deposition to high Arctic snow, Atmos. Chem. Phys., doi:10.5194/acp-17-5775-2017, 2017. 2/15 Referee Comment: Please clarify the phrase "less prone to the ambiguities introduced by snow-pack collection". Response: A comparison of fresh and aged snow sampling was discussed in the previous paper. However, we agree that this line should not be included here without additional clarification or references. In the interest of space, this line has been removed from the revised manuscript. 2/27-28 Referee Comment: Mention measurements data here – otherwise it sounds like the study

includes only PMF and air mass modeling. Original Line: In this context, this paper analyses the sources of chemical components in freshly-fallen snow samples collected over a complete fall-winter-spring at a high Arctic location (Alert, Nunavut), using a combination of Positive Matrix Factorization diagnostics and Lagrangian dispersion modelling. Response: The line has been revised as suggested: "In this context, this paper analyses the sources of chemical components in freshly-fallen snow samples collected over a complete fall-winter-spring at a high Arctic location (Alert, Nunavut) and analysed for a broad suite of analytes, using a combination of Positive Matrix Factorization diagnostics and Lagrangian dispersion modelling." (3/4-7) 7/3 Referee Comment: Is this supposed to be 59 samples (based on P3 L2)? Response: The referee is correct that a total of 59 sets of samples were analysed in the course of this study; however, some collection periods did not provide sufficient snow volume to perform the complete suite of analyses (see referenced Macdonald et al., 2017 for the complete list of sampling dates, completed analyses, and results). Section 2.4.1 of the manuscript explains that the PMF analysis was limited to collection periods with the majority of analytes of interest measured (original manuscript 5/21; revised manuscript 5/12). 7/4 Referee Comment: Clarify wording that you are discussing analyte concentrations and fluxes. Original Line: Three metrics were considered as the basis for this analysis: snow concentration, flux per day, and flux per snowfall (i.e., assuming each sample represented a single snowfall event regardless of the time period over which it occurred, which is known to be true for the majority of samples based on Alert station operator records). Response: The line has been revised to clarify as follows: Three metrics were considered as the basis for this analysis: analyte concentration, flux per day, and flux per snowfall (i.e., assuming each sample represented a single snowfall event regardless of the time period over which it occurred, which is known to be true for the majority of samples based on Alert station operator records). Per the referees' suggestions, details on the PMF analysis have been largely moved to the supplemental, including this line. (revised supplemental page 10) 7/13-14 Referee Comment: This discussion is not intuitive and could be clarified further. Can we learn about processes from these differences? Original Line: The source contributions identified by the flux per snowfall period analysis were the most readily interpreted as physically realistic factors. Moreover, this metric showed the largest correlation between BC snow and atmospheric measurements (Pearson's correlation coefficients of 0.4, 0.3, and 0.5 for BC concentration, flux per day, and flux per snowfall period, respectively), implying that the flux per snowfall may in general be more closely related to the change in analyte sources over time while concentration and flux per day may be more intrinsically dependent on changes in deposition processes. Response: Per the referee comments, discussion of the concentration and flux/day PMF analyses has been moved from the manuscript to the supplemental. The following has been added to this discussion in the supplemental to clarify: "For example flux per snowfall is likely related to a specific synoptic event, arising from a common location. This will be more useful than concentration given that this value will be affected by the amount of precipitation, and more useful than flux per day that will be affected by the rapidity of snowfall." (revised supplemental page 10-11) 7/17-33, 8/5-9, and Table 1 Referee Comment: 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. Response: Per the referee's suggestion, this section has been shortened in the manuscript. Table 1 has been left in the manuscript as it lists the analytes included in the PMF analysis and specifically which were considered strong or weak. We agree that the diagnostic properties are not necessary in the manuscript for the target audience of the ACP; however, we do believe it is important to list the analysis main inputs and describe the overall fit of the predicted results. 8/14-15 Referee Comment: These sentences are redundant. Original Line: These compounds are all typical of sea salt, suggesting a marine origin for Factor 1. The composition of Factor 1 was found to be consistent with that of sea salt (Pytkowicz and Kester, 1971). Response:

The intention of these two lines was to convey that the dominant compounds as well as their relative proportions were both consistent with a marine source. We agree that as written this distinction is not clear and the lines become redundant. The line has been revised as follows: These dominant analytes and their relative proportions are consistent with that of sea salt (Pytkowicz and Kester, 1971), suggesting a marine origin for Factor 1. (11/10-11) 9/1-12 Referee Comment: "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.) Response: The word "compounds" has been removed, and typically replaced with "analytes" as to be general. The uncertainty of enrichment ratios have been described in the text using the 25th and 75th bootstrapping analysis results. (11/8-12/3) Figure 1 Referee Comment: Remove "(point)" and "(bar)" on the y axes, as this is already shown in the legend, and "bar" is a unit of pressure. Response: Addressed in revised manuscript. Figure 1 Caption Referee Comment: Provide further description of how to interpret the figure for improved clarity, particularly for those not familiar with PMF. 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 authors recognize 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. (revised page 8) Figure 2 Caption Referee Comment: What are the traces normalized to (themselves, other factors?)? What are the units? This caption is not clear. Response: Normalized factor contribution is the metric provided directly by the EPA PMF analysis and is the typical method used to discuss these results. A thorough discussion of this metric and its interpretation are provided in the EPA PMF guide (Norris et al., 2014). However, the authors recognize that not all readers will be familiar with such analyses. For clarity, the Figure 2 caption has been revised as follows: Normalized factor contribution. The unitless contributions describe the relative magnitude of each factor over time such that the average contribution of each factor is one. (revised page 9) 11/9 Referee Comment: Provide the calculated ratio in parentheses for context. Original Line: Specifically, the modelled ratio of As/Al was seen to be closer to that of local soils (Barrie, den Hartog, and Bottenheim, 1989) than the global typical composition (Taylor, 1964; Masson-Delmotte et al., 2013) with enrichment ratios of 6 and 37, respectively. Response: This primary focus of this line is to convey the greater similarity of the apportioned factor to local soil as compared to typical global soils. This is exemplified with the enrichment ratios provided. The line has been revised to also provide the ratios as follows: Specifically, the modelled ratio of As/Al (0.00081 m/m) was seen to be closer to that of local soils (0.00013) (Barrie, den Hartog, and Bottenheim, 1989) than the global typical composition (0.00002) (Taylor, 1964; Masson-Delmotte et al., 2013) with enrichment ratios of 6 and 37, respectively (6.3-9.5 and 37-58 25th-75th percentiles per bootstrapping analysis). (12/28-31) 13/32 Referee Comment: 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. Response: The updated manuscript provides all neutralization ratios in the revised Table 2. It is clarified that the provided formula is used for all calculations. (9/8-11) Table 3 Referee Comment: 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. Response: We agree that the original naming could be misconstrued. This has been revised as “Open Ocean”. (revised page 11) Figure 4 Referee Comment: The abbreviation “Cbx. Ac.” In the legend is not immediately obvious; I suggested writing out “carboxylic acids” on two lines instead for improved clarity. Response: Figure revised with “carboxylic acid” as legend entry. (revised page 10)

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-718/acp-2017-718-AC2-supplement.pdf

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