Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-944-AC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations of Atmospheric Chemical Deposition to High Arctic Snow" *by* Katrina M. Macdonald et al.

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Received and published: 27 February 2017

Referee comments received and published: 6 December 2016 (quoted below)

We thank Referee #1 for their well-thought comments and constructive criticism. The following section recapitulates the referee's comments and provides the authors' response to each point presented by the referee.

Response to Referee Discussion

Referee Comment: This manuscript presents results from a 9-month long campaign that collected samples of freshly fallen snow as soon as possible after each deposition event that occurred at Alert from mid-September 2014 through mid-May 2015. These samples were obtained using a technique that the authors assert allowed estimation of

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Discussion paper

combined wet and dry deposition over the interval between each collection, and were analyzed for BC, major ions, and a large number of trace elements. The authors also use measurements of the concentration of aerosol-associated BC, ions and a smaller number of trace elements made as part of the baseline suite by station staff to explore the transfer of impurities from the atmosphere to the snow.

Response: We thank the reviewer for this summary.

Referee Comment: The data set is quite rich and interesting, however I feel that the interpretation and synthesis does not exploit the data set well. A major problem is the decision to define and rely on the “effective deposition velocity” (EDV) for nearly all of the discussion. The authors acknowledge on page 2 that wet deposition is dependent on a wide range of factors, many of which change from day to day, such that previous theories have never found it useful to consider a wet deposition velocity. Then on page 9 they point out additional reasons that such a construct is bound to obscure critical details relating to the processes removing aerosol associated (and gas phase) impurities from the atmosphere to the snow surface, without presenting a strong argument why ignoring these details is reasonable.

Response: We agree with the reviewer that an "effective deposition velocity" lumps together both dry and wet deposition, and that this term is not commonly used in the literature. We made this point clearly in the original paper, as pointed out by the reviewer. The reason we chose this approach was simply that it is not possible to accurately separate dry and wet deposition via snow collection/analysis of the type performed in this study. However, discussion of this parameter does provide insight on bulk deposition mechanisms and magnitudes, and indicate when these exceed that expected due to dry deposition alone. Thus, the effective deposition velocity provides a holistic view of how deposition processes change over the winter seasons. As well, the "effective deposition velocity" is a quantity that can be readily extracted from GCM or CTM models, for comparison to the data. Section 3.2.1 has been expanded to give greater clarity on the interpretation and use of this parameter. We thank the reviewer for this comment.

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Discussion paper

Referee Comment: To my mind this bad decision is compounded by focusing on monthly values of the estimated EDV as the highest temporal resolution (but only shown in Figure S3). In the actual manuscript one of the key figures (Fig. 2) shows estimated EDV for BC and ions, but only as box and whisker plots summarizing the full campaign (minus Jan and Feb which were discarded due to concerns about data quality, more on that later). Another key figure (Fig. 4) shows monthly EDV, but lumps all of the different impurities together, obscuring significant differences that are evident in Fig. S3 with no explanation. Likewise, Fig. 5 does include separate EDVs for BC and the ions, but presents statistical summaries for a 3 month warm season compared to 4 month cold season (again, omitting Jan and Feb).

Response: We agree with the referee that higher resolution effective deposition velocities should be included to allow inferences of specific events. However, we believe monthly values allow better insight into the general deposition regime of Arctic aerosol without the complexity of event-to-event variation. The supplemental Figure S3 has been moved to the manuscript as per the suggestion of both referees, now Figure 2. Six-day values have been added to this figure along with their associated uncertainties. Monthly values are still included for the discussion of variance in deposition by composition and temporally. Section 3.2.2 of the manuscript has been revised as follows to address this issue (Page/line in the revised text: 10/12-23):

Revised text: Monthly effective deposition velocities were used to contrast deposition mechanisms by aerosol composition. A monthly resolution provides insight into the general deposition regime of each analyte, highlighting the impact of bulk deposition characteristics rather than event-specific variability. The variability between aerosol of different composition and the influences of seasonal changes within the Arctic system are simpler to identify without the interference of variability between event-specific conditions. A monthly analysis also facilitates future comparison with modelled results which may not replicate individual events. January and February, 2015, were excluded from the monthly analysis because blizzard and high wind conditions were believed

to have caused significant losses of snow from the snow tables during these months (based on operator reports), which would lead to underestimation of these snow flux values. The effective deposition velocity is best suited to analysis across periods of equal length and precipitation volume, since both of these parameters are inherently included when the wet deposition efficiency is converted to an equivalent deposition velocity. With the exception of January and February, the total monthly snow precipitation over the campaign was relatively constant, with a relative standard deviation of 20%.

Referee Comment: The authors should establish a stronger case for the utility of the EVD, and it should be calculated and presented at the 6-8 day resolution of the aerosol sampling. A revised version of Fig S3 (with approximately 4 times as many data points) should be in the main text, informing most or all of the discussion. In the detailed comments below I will work through the manuscript page by page with a mixture of editorial and content comments. The latter will be based on statements in this draft in relation to the figures that are now included, hence may be less relevant if a revised manuscript focuses on the higher resolution that the data set could provide.

Response: Please see our comments above.

Referee Comment: Before turning to detailed comments I need to raise one other general objection to this draft. The authors claim repeatedly that this data set is unique by virtue of the large number of impurities quantified, that their sampling allowed fluxes to be determined, and that sampling captured all (and only) fresh snow events. I would like to point out that the DGASP campaign at Dye 3 in south Greenland took a very similar approach back in 1988-89, though many of the snow samples were not collected in a way that allowed net accumulation (hence flux) to be quantified. This shortcoming of DGASP sampling was addressed in a long series of summer time campaigns at Summit, Greenland (1989 to at least 2003) that included daily sampling to quantify both concentration and the mass of impurity/unit area in the surface layer of snow (see series of papers by Bergin et al. from the mid 90's with "Flux" in their titles as

examples), and was maintained for year round campaigns at Summit in 1997-1998 and 2000-2002 (Dibb et al., 2007). With better hindsight, the authors may want to rephrase the statements in this draft made on page 1, lines 20-21; page 2, lines 1-2; page 3, lines 2-3; page 3, lines 20-22; page 6, line 12; page 7, lines 3-6 (particularly this one!); page 16, lines 3-5.

Response: Referee #1 referred to two studies which provided similar insight to Arctic snow deposition and should be discussed: the Dye 3 gas and aerosol sampling program (DGASP), which collected atmospheric and snow samples in Greenland between August 1988 and July 1989 (Davidson et al. 1993; Jaffrezo, 1993); and the snow flux papers published by Bergin et al. in the mid 1990's, which consisted of two month-long campaigns collecting snow, fog, and dry deposition in Greenland in the summers of 1992 and 1993 (Bergin et al., 1994; Bergin et al., 1995).

The DGASP study was an excellent campaign with interesting results. Specifically, surface snow samples were collected at Dye 3 following each snow fall during the 1988-89 season, conditions allowing. Samples were analyzed for major ions, metals, optical carbon, and particle size distribution (Jaffrezo, 1993). As mentioned by Referee #1, the deposition area was not measured for all DGASP samples; thus, snow flux could not be calculated. Another distinction between the DGASP campaign and this study is the collection strategy: surface snow collection as compared to snow table collection as per this study. Collection of snow samples from the ground introduces the inherent difficulty of determining the appropriate collection depth and distinguishing fresh and aged snow. Snow table collection avoids this concern; however, both techniques are subject to uncertainty due to redistribution of snow by winds. Thus, the deposition area and time period associated with surface samples may be less certain than that those associated with snow table samples. Furthermore, the collection of dry deposition from between snowfalls would likely differ between the techniques. As mentioned in the paper, it is uncertain what portion of this dry deposition would have been collected from the snow table; however, we believe that this collection efficiency would be similar between

samples. The collection of such dry deposited material in surface snow samples is even harder to quantify, but would be very dependent on small deviations in the collection depth. Thus, we believe the snow table collection technique distinguishes this paper in its ability to quantify the bulk deposition of chemical species as a flux.

The Bergin et al. papers discussed by the referee are also wonderful studies with valuable insight on deposition within the Arctic. These studies quantified daily snow, fog, and dry deposition to collection surfaces at Summit, Greenland during the summers of 1992 and 1993. Samples were analysed for major ions and particle size. The known collection area and time associated with these samples allowed calculation of the material flux and deposition velocity. The Bergin measurements make an excellent complement to the data of this study as they focus on different seasons (summer as compared to fall/winter/spring). Although the measurements of this study are less appropriate for distinguishing the exact split between deposition mechanisms, they do represent a longer collection campaign and broader suite of analytes. As discussed by Bergin et al. (1995), long-term studies are valuable for accessing the annual inventory of deposition within the Arctic.

References to the DGASP and Bergin et al. studies have been added within the introduction, methodology, and results sections (see specific instances of note below). We apologize for not having these in the paper from the outset.

Bergin, M. H., Jaffrezo, J. L., Davidson, C. I., Caldow, R., and Dibb, J.: Fluxes of chemical species to the Greenland ice sheet at Summit by fog and dry deposition, Geochim. Cosmochim. Ac., 58 (15), 3207–3215, doi:10.1016/0016-7037(94)90048-5, 1994.

Bergin, M. H., Jaffrezo, J., Davidson, C. I., Dibb, J. E., Hillamo, R., Maenhaut, M., Kuhns, H. D., and Makela, T.: The Contributions of snow, fog, and dry deposition to the summer flux of anions and cations at Summit, Greenland, J. Geophys. Res., 100 (D8), 16275–16288, doi:10.1029/95JD01267, 1995.

Davidson, C.I., Jaffrezo, J. L., Mosher, B.W., Dibb, J.E., Borys, R.D., Bodhaine, B. A., Rasmussen, R. A., et al.: Chemical constituents in the air and snow at Dye 3, Greenland âĂŤ I. Seasonal Variations, Atmos. Environ. A-Gen., 27 (17), 2709–2722, doi:10.1016/0960-1686(93)90304-H, 1993.

Jaffrezo, J. L. and Davidson, C. I.: The Dye 3 gas and aerosol sampling program (DGASP): An Overview, Atmos. Environ. A-Gen., 27 (17), 2703–2707, doi:10.1016/0960-1686(93)90303-G, 1993.

Specific lines discussed by the referee are listed below with the revised text by Page/Line(s) #.

1/20-21

Original Line: These measurements are a unique data set for comparison to models that incorporate deposition to high Arctic snow.

Revised Line: Line removed from updated text.

2/1-2

Original Line: Observations of fresh snow samples are particularly uncommon and previous explorations of snow deposition and scavenging mechanisms have been largely reliant on aged snowpack sampling, modelling, and laboratory tests.

Revised Line: Seasonal observations of fresh snow samples are particularly uncommon (e.g., Davidson et al., 1993; Toom-Sauntry and Barrie, 2002; Hagler et al., 2007) and previous explorations of snow deposition and scavenging mechanisms have been largely reliant on short-term or aged snowpack sampling (e.g., Bergin et al., 1995), ice cores (e.g., Legrand and De Angelis 1995), modelling, and laboratory tests. (2/2-5)

3/2-3

Original Line: To our knowledge, this is the first time that the composition of freshly fallen snow has been analyzed at high temporal frequency throughout an entire cold

season in the high Arctic.

Revised Line: To our knowledge, this is the first time that the composition and flux of freshly fallen snow has been analyzed at high temporal frequency throughout an entire cold season in the high Arctic. (3/4-5)

3/20-22

Original Line: This ability to assign a precise deposition area and time period to each sample, minimizing the inaccuracies introduced by extensive snow redistribution by winds or snow sublimation/melt, was a considerable advantage over previous sampling campaigns of aged snowpack.

Revised Line: The collection of fresh snow samples reduces the impact of snow sublimation and/or melt as well as the movement of chemical species between snow and air, which can be a concern for snowpack sampling; however, some bi-directional exchange between snow and atmosphere is unavoidable and natural within natural snowpack systems. Also, the collection of samples from a snow table eliminated the need to estimate the depth of fresh snow, a source of uncertainty for traditional surface snow sampling. This ability to assign a well-defined deposition area and time period to each sample was an advantage over traditional sampling campaigns of aged snowpack . However, both this and traditional snow collection techniques are prone to the uncertainty introduced by the redistribution of snow by winds. Measurements of snowfall accumulation were not available for the collection site. Snow depths measured at the Alert ECCC station indicate that the snow collected on the tables may have underestimated the total snowfall volume by a factor of approximately 1 to 10; however, the meteorological station and collection site were separated by over 6 km with a 50 m difference in elevation, and there was significant disagreement between operator records of weather and that indicated by the meteorological station (see supplemental S4.2 for details). Thus, it was unclear whether this disagreement was the result of snow loss from the snow table or the natural spatial variability in precipitation, and no correction

was applied to the collected snow depth. (3/28-4/8)

6/12

Original Line: Unlike traditional snowpack collection campaigns, each sample for this study was collected fresh after a known time and over a known area.

Revised Line: Each sample for this study was collected fresh after a known time and over a known area. (6/15)

7/3-6

Original Line: Comparison of the measured mean values with previous snow observations by others provided a general corroboration of the measurements (see supplemental Table S7 for details); however, given the limitations of previous snow measurements as discussed above, no extensive comparison of seasonal trends could be conducted.

Revised Line: A review of existing Arctic snow measurements found the measured median mixing ratios to fall within expected ranges (see supplemental Table S7 for details); however, it should be noted that the referred data represent a variety of collection and analysis techniques. In general, measurements of this campaign showed salt species and non-crustal metals to be at the lower end of the typical range while SO42- and NO3- were at the higher end. A limited number of seasonal snow collection campaigns were available for comparison of the observed seasonal trend in analytes. The winter peak in BC was similar to that observed by Davidson et al. (1993); however, spring values observed in this campaign were higher than previously seen. The observed seasonal trend in major ions was generally consistent with existing literature (Davidson et al. 1993; Toom-Sauntry and Barrie, 2002). Specifically, a winter peak in sea salt, fall/spring peaks in MSA, and a winter peak in NO3- are all typical. However, the fall peak observed in the NSS-SO42- mixing ratio and spring peak in NO3- were unlike seasonal trends observed previously (Davidson et al. 1993; Toom-Sauntry and Barrie, 2002). (9/1-10)

16/3-5

Original Line: Given the rarity of temporally-refined and broadly speciated Arctic snow sampling campaigns, measured deposition magnitudes and insights on deposition mechanisms such as these are valuable for future model validation.

Revised Line: We have left this line as is. Although the referee has provided some additional references, we still find such campaigns to be uncommon. No similar campaign has been completed in the recent decade.

Response to Detailed Comments

Referenced to Page/Line #(s) in the original manuscript:

1/16

Referee Comment: as noted above "effective deposition velocities" not established, and maybe not useful, a strong case needs to be made later that the concept does have utility, or these calculated values should not be highlighted in the abstract

Original Line: Comparison with simultaneous measurements of atmospheric aerosol mass loadings yields effective deposition velocities that encompass all processes by which the atmospheric species are transferred to the snow.

Response: As mentioned above, the analysis of effective deposition velocity provides insight into the general deposition regime of the measured analytes which guides the discussion of this paper. Furthermore, this quantity can be directly compared to GCM and CTM output. Because such models cannot often model individual synoptic events, a monthly averaged value is likely of more utility than individual values alone. That said, the individual measurements are now also included in the paper.

2/25

Referee Comment: dependent on snow type

Response: Editorial comment corrected in text (revised manuscript page/line 2/28).

2/27-29

Referee Comment: not sure what the message of sentence starting “The efficiencies...” is supposed to be Original Line: The efficiencies of below-cloud scavenging of gases and particles can be similar at an order of magnitude level, though measurements of gaseous scavenging in particular are infrequent and dependent on composition (McMahon and Denison, 1979).

Response: This line has been removed from the revised manuscript.

2/30

Referee Comment: Last sentence is quite true, and for pretty good reasons. Why are you bucking the trend?

Original Line: Hence, wet deposition is not typically described via a deposition velocity.
Response: As discussed above, the methodology of this study did not allow for accurate split of dry and wet deposition. Thus, a bulk parameter has been presented which allows us to explore the general deposition regime of Arctic aerosols by composition and season. This bulk parameter does provide insight into the degree to which the aerosol transport into snow exceeded that expected due to dry deposition alone.

3/11

Referee Comment: Based on the picture in Fig S2 I strongly suspect that the snow table had a major problem with undercatch if there was any wind at all (not just high winds causing blowing snow). It would have been prudent to check this fairly frequently through the campaign by determining the cumulative accumulation of snow on the ground near the table. At a minimum, determination of SWE from either a small pit or a simple core could have been compared to a summation of the SWE for all of the layers collected off the table. Better would have been to check the inventory (mass/unit area) from snow surface to the ground of selected impurities at some interval (at least

monthly), by collecting and analyzing the full column of accumulated snow. In the absence of this kind of validation, you probably need to increase the uncertainty estimate for your impurity fluxes.

Response: These are good points that have now been raised as uncertainties in the paper, in section 2.1.

4/16

Referee Comment: What were the QC runs?

Response: Quality control runs were completed by re-analyzing the calibration solution every set number of samples to quantify any instrument drift. An extended supplemental has been prepared with additional details on the methodology. This includes a description of the QC runs for each analysis in supplemental section S2.2.

4/31

Referee Comment: How, exactly, was "mass recovery precision" determined? Sounds like some kind of yield tracer was added, but no details given.

Response: Mass recovery was quantified by massing the total sample at several stages over the sample preparation and analysis. This was done to confirm no sample was lost throughout handling. An extended supplemental has been prepared with additional details on the methodology, including the following text: "An average sample mass closure of $\pm$1% was observed over the digestion procedure (i.e., sample mass was measured at all stages of analysis to confirm significant sample was not lost during handling)." (supplemental section S2.1.3).

5/4-6

Referee Comment: When was this "internal standard" added? Before or after digestion? Did it also serve as the yield tracer to track recovery?

Response: The internal standard was added to the digested solution during analysis.

An extended supplemental has been prepared with additional details on the methodology, including the following text: “An internal standard was included in the ICP-MS analysis to quantify and correct for any instrument drift or inter-sample variability. The SCP Science Int. Std. Mix 1 was selected so as to minimize interference with measured analytes while covering the full analyzed spectrum of mass to charge ratios. This internal standard was added to the digested solution as they were sampled for analysis.” (supplemental section S2.2)

5/11

Referee Comment: in a class 100

Response: Editorial comment corrected in text (5/10).

5/25-26

Referee Comment: Are U. Toronto MA thesis publicly available? If not, are they citable in ACP?

Response: Pertinent details of this thesis have been moved into the supplemental, section S2, and the reference removed.

6/14-24

Referee Comment: There is a sizeable literature about the problems with surrogate surfaces for the collection of both dry and wet deposition. Some of that should be cited here.

Response: We agree that additional references should be provided here and apologise for not doing so. The following have been added (6/18-19):

Davidson, C. I., Lindberg, S. E., Schmidt, J. A., Cartwright, L. G., and Landis, L. R.: Dry deposition of sulfate onto surrogate surfaces, J. Geophys. Res., 90 (D1), 2123–30, doi:10.1029/JD090iD01p02123, 1985.

Hicks, B. B.: Measuring dry deposition: A Re-assessment of the state of the art, Water Air Soil Poll., 30, 75–90, doi:10.1007/BF00305177, 1986.

Ibrahim, M., Barrie, L. A., and Fanaki, F.: An Experimental and theoretical investigation of the dry deposition of particles to snow, pine trees and artificial collectors, Atmos. Env., 17 (4), 781–88, doi:10.1016/0004-6981(83)90427-4. 1983.

6/24-25

Referee Comment: May want to include comparison between cumulative depth, or cumulative SWE probably better, on the table and at the met station. This would allow the reader some insight into how bad table undercatch may have been (or persuade you to make some correction).

Response: While we agree that the depth of snow captured by the snow tables was likely impacted by winds, we do not believe the local meteorological station provides a better estimate of the actual precipitation depth. The Alert ECCC meteorological stations are over 6 km away with a 50 m difference in elevation. Such a difference in location could lead to significant differences in observed snowfall. Furthermore, observations of the collection station operators showed many instances of disagreement with the meteorological station record. For example, there were periods when the operators noted snowfall and yet the meteorological station saw none, or vice-versa. There were also times when the operators noted blowing snow, yet the meteorological station did not indicate this. Thus, we do not believe that the snow depth measured by the station would be an appropriate comparison to that seen at the collection site. Please see revised line of 3/20-22 above.

However, we have performed a supplementary analysis with the assumption that the snow depths measured at the meteorological station are applicable to the snow collection site. These adjusted median effective deposition velocities ranged from 0.08 to 1.7 cm/s, as compared to 0.03 to 1.1 cm/s using snow table depths. The relative velocities and interpretation of the deposition characteristics remained unchanged.

6/27

Referee Comment: Flux per day over 2-18 days makes no sense for a wet deposition event likely lasting just hours. Understood that you cannot separate wet and dry in this data set, but this contributes to discomfort with the calculated EVDs.

Response: The referee is quite right that the deposition measured does not relate to that of a single snowfall but total deposition over a period. As per the discussion above about the utility of the effective deposition velocity, total flux over a period provides insight into the bulk movement of material in the Arctic atmosphere.

8 (Fig 1)

Referee Comment: Why are there gaps of different lengths and at different times in the time series of BC, ions, and crustal metals? According to section 2.1, anytime one bottle was filled there should have been 3 others (presumably for the different analyses). Problems causing gaps need to be noted and enumerated (probably better in 2.1 than here).

Response: There were events when insufficient snow volume fell to fill all sample bottles. In these cases a sub-set of the samples was collected. The samples collected on each date are listed in Table S1. The following text has been added to section 2.1 "When insufficient snow volume was available for complete collection, a subset of the replicate samples was collected as listed in supplemental Table S1." (3/20-21).

9/13-16

Referee Comment: Why are the much more numerous flights from the 4 NOAA AGASP campaigns or the 5-6 NSF TOPSE flights not included here? My impression is that some of these showed significant gradients in the lowest 3 km. (Note that the special issue of Atmos Environ on DGASP also includes AGASP3 papers and provides access to the earlier ones.)

Response: Although some previous studies have shown the lower Arctic atmosphere

to be somewhat more consistent, upon further investigation we agree with the referee that this is not always the case. Therefore, we have rephrased this section as follows to discuss this limitation: “A caveat to this analysis is that the three deposition mechanisms relate to different atmospheric concentrations, a gradient which is not necessarily captured when the ground-level atmospheric concentration (CA) is used to calculate the effective deposition velocity: dry deposition affects the lower atmosphere, in-cloud scavenging the cloud layer, and below-cloud scavenging the full below-cloud atmospheric column. Previous observations of vertical profiles in the Arctic have shown notable variability with altitude (Hansen and Rosen, 1984; Leaitch et al., 1989; Spackman et al., 2010; Brock et al., 2011; Sharma et al., 2013). So, the calculated effective velocity includes an intrinsic variability dependent on the vertical atmospheric profile of each analyte.” (9/30-10/4).

Brock C. A., Cozic, J., Bahreini, R., Froyd, K. D., Middlebrook, A. M., McComiskey, A., Brioude, J., et al.: Characteristics, sources, and transport of aerosols measured in spring 2008 during the aerosol, radiation, and cloud processes affecting Arctic Climate (ARCPAC) Project; Atmos. Chem. Phys., 11, 2423–2453, doi:10.5194/acp-11-2423-2011, 2011.

Hansen, A. D. A., and Rosen, H.: Vertical distributions of particulate carbon, sulfur, and bromine in the Arctic haze and comparison with ground-level measurements at Barrow, Alaska, Geophys. Res. Lett., 11 (5), 381–84, doi:10.1029/GL011i005p00381, 1984.

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.

Leaitch, W. R., Hoff, R. M., and MacPherson, J. I.: Airborne and lidar measurements of aerosol and cloud particles in the troposphere over Alert Canada in April 1986, J. Atmos. Chem., 9, 187–211, doi:10.1007/BF00052832, 1989.

Spackman, J. R., Gao, R. S., Neff, W. D., Schwarz, J. P., Watts, L. A., Fahey, D. W., Holloway, J. S., et al.: Aircraft observations of enhancement and depletion of black carbon mass in the springtime Arctic, Atmos. Chem. Phys., 10, 9667–9680, doi:10.5194/acp-10-9667-2010, 2010.

9/16-18

Referee Comment: Somewhat bad form to use unpublished data to support your position, but beyond that, flights in April are not really informative about stability and stratification in the winter. Recall, you are asserting these data are unique because they include the cold season (see later comments related to your Figs 4 and 5).

Response: We agree with the referee that this is a very good point. The reference to unpublished data has been removed. The discussion of vertical variability has been revised as described in the previous point.

9/21-27

Referee Comment: As noted, assuming you can make a better case for utility of EVDs, I do not like the use of monthly averages here and do not think the final sentence is valid justification. There are a lot of papers focused on the POLARCAT airborne campaigns where global and regional CTMs attempted to simulate day by day, plume by plume in some cases, and more should be expected as part of recent and future campaigns. Modelers are striving to improve their simulations, so if experimentalists provide high resolution records they welcome the challenge of simulating them, however poorly the first round of comparisons turn out.

Response: Yes, we agree that some models are capable of high-resolution results; however, monthly values would be of use to a variety of lower-resolution models and seasonal trends can be compared across models simulating different years. Please refer to response above discussing the use of monthly values within the paper's discussion.

9/28

Referee Comment: As noted earlier, summarizing 7 months in box and whisker plot makes little sense to me.

Response: Please see response above.

10/10-13

Referee Comment: To me, the most puzzling aspect of Fig 2 is why Ca is so much higher than Na, Cl and Mg. This is not even mentioned until page 15 (with no plausible explanation). Similarly, suggesting here that BC, NH4 and SO4 seem dominated by dry dep causes some tension with later suggestion that they are serving as CCN (or at least being efficiently scavenged) in S/O/My (Fig 5).

Response: Discussion of the high Ca velocity has been moved to this section (13/15-17). As mentioned in the original manuscript, we do not have sufficient information to make a definite claim as to the processes resulting in the elevated Ca deposition relative to Mg. However, the previous study quoted in the text (Banta et al., 2008) showed that distinct Ca-rich and Mg-rich crustal particles may typically exist within the Arctic. Therefore, we suggest that the discrepancy is a result of differing properties associated with these different crustal sources.

We show that the effective deposition velocities of BC, NH4+, and SO42- over the colder months fall within the typical range of dry deposition velocities for accumulation mode particles. This indicates that dry deposition is a dominant deposition mechanism for these chemical species over cold months. Within the warmer months, the effective deposition velocities of BC, NH4+, and SO42- are still relatively low but do show some enhancement above the typical dry deposition range. Thus, we suggest that they experience enhanced contribution from wet deposition over this period. Specifically, this enhance wet deposition may be related to their activity as CCN. However, dry deposition is likely still a significant factor for these species over warm months as well. We do

not believe these statements are contradictive.

11/5-6

Referee Comment: Not so sure this is true. The Hi-Vol sampling at Alert used to use cellulose filters (Whatman 41s) which do collect acids with decent efficiency. And for a filter exposed for 6-8 days, seasalt and dust can build up to the point where acids may be collected quantitatively.

Response: The Hi-Vol collection was completed using cellulose Whatman 41 filters, as has been the procedure at Alert for decades (Barrie and Hoff, 1985). We agree that although these filters are intended to sample particles they are subject to both positive and negative artefacts. Gases may adsorb onto the filter or acidic gases may react with alkaline sea salt or dust, if they are present, and thus become part of the sample. Gases may also re-volatilize due the high sample flow, or particles may react with acidic Sulphur species on the filter and be released as a gas. However, given the observed chloride deficit in the Hi-Vol aerosol sample, we suspect that the collection of gaseous species by Hi-Vol sampling was likely not a major contribution.

The text has been revised to clarify that Hi-Vol measurements are likely dominated by particle collection, though not necessarily absent of gaseous aerosol collection. Original Line: Due to the nature of the Hi-Vol analysis, the measured atmospheric concentrations include only the particle-phase ambient species; however, snow can scavenge both the particle and gas phases, and thus snow samples provide a composite measurement of both. Revised Line: It is expected that the Hi-Vol measurement technique would collect predominantly atmospheric particles, while snow would scavenge both gaseous and particulate aerosol (12/17-18). Barrie, L. A., and Hoff, R.M.: Five years of air chemistry observations in the Canadian Arctic, Atmos. Environ., 19 (12), 1995–2010, doi:10.1016/0004-6981(85)90108-8, 1985.

10/7 to 11/8

Referee Comment: Chloride deficit in MBL aerosol and excess in Arctic snow (especially in summer) has been well documented by many others in addition to Toom-Sauntry and Barrie, 2002. This section seems too long for significance of the point being made, but also lacking references to prior work.

Response: We agree that this section need not include such a detailed discussion of chloride deficit. The bulk of this discussion and associated plot have been moved to the supplemental. Furthermore, additional references have been added (12/11):

Barrie, L. A., and Hoff, R.M.: Five years of air chemistry observations in the Canadian Arctic, Atmos. Environ., 19 (12), 1995–2010, doi:10.1016/0004-6981(85)90108-8, 1985.

Quinn, P. K., Bates, T. S., Schulz, K., and Shaw, G. E.: Decadal trends in aerosol chemical composition at Barrow, AK: 1976–2008, Atmos. Chem. Phys., 9, 18727–43, doi:10.5194/acpd-9-18727-2009, 2009.

12/14

Referee Comment: enhanced deposition

Response: Editorial comment corrected in text (13/12).

12/18-21

Referee Comment: As pointed out in first sentence of this paragraph, everything shown in Fig 2 has higher EVD than BC, so what is so special about these 4? They are not as enhanced as Ca in the median, and max EVD for K, Mg, C2O4 and Ca are all higher than the max for MSA, Br and Cl. I agree that the 4 you highlight can have appreciable gas phase, but that is not something discovered or even really supported by Fig 2.

Original Line: Gas-phase deposition to snow is suggested to contribute to the observed enhanced velocities of methanesulphonate (MSA), Br, Cl, and NO3 relative to BC, either due to gas-phase emissions or gas-phase partitioning during aerosol aging

followed by subsequent deposition.

Response: As stated by the referee these four analytes were specified as they can all have appreciable gas phase. We do not claim to have discovered this point, and believe the text reflects this. Rather we have simply shown that our observations are consistent with this and that the gas-phase deposition can be an important factor in the bulk deposition of chemical species within the Arctic.

Pages 12-14 (Figs 4 and 5)

Referee Comment: As noted at top, I like that 4 at least shows monthly resolution, but don't like lumping all impurities, while 5 is nice in showing BC and all ions, but is averaging over too much time.

More importantly, I think both are missing key point (which calls the value of EVD into question). Perhaps the most important feature of the Arctic is the huge seasonal contrast in daylight, which leads to big contrasts in energy balance and atmospheric stability. In the winter the atmosphere is extremely stable with weak vertical mixing. Atmospheric impurities that get into the BL tend to be trapped there, and concentrations can be high. As things warm up the BL deepens and concentrations tend to decrease close to the surface. This is clearly reflected in Table S8 which shows very low concentrations of BC, Cl, C2O4, Na, K, Mg in SOMy compared to the colder months. Pattern is not so strong for NO3, NH4 and SO4, but the averages for the 3 warm months are quite low compared to the 6 (or 4) cold ones. Part of this may be transport related, and local biogenic emissions may account for the weaker trend in SO4 and opposite trend in MSA, but dilution into a much deeper BL is probably an important factor.

As a thought experiment, given constant monthly accumulation of snow, and constant burden of impurities in the BL, and assuming complete cleansing of the BL over each month, but a 5-10 fold deeper and well mixed BL in warm months compared to cold ones, what would the EVDs look like. Constant monthly flux to snow divided by 5 (or 10) fold lower aerosol concentration measured at surface would yield 5 (or 10) times larger

EVD in the warm months. Clearly, the story is not so simple. But, is it really plausible that scavenging in mixed phase clouds is the major factor accounting for increase in BC and all the ions (which should all behave differently in cloud)?

See again the suggestion to include a higher resolution version Fig S3 as the focus of discussion, and exploit the differences to gain insight.

Response: The focus of our discussion is the seasonal variation in bulk deposition processes. We agree that there is significant and interesting variability in deposition at an event resolution; however, the influences on individual events are very complex and cannot be accurately judged without additional information. A holistic view of the scavenging and deposition process can be obtained through analysis and discussion of monthly values to provide insight into their gross mechanisms and better differentiate seasonal trends from the random variability between snowfall events. We have added high-resolution data to the manuscript to guide discussion and future research, but have kept seasonal variation as the focus of our analysis.

We agree that the boundary layer height could be an influence on deposition, as stated by the referee. We have obtained radiosonde measurements from Alert to better characterize the vertical profile. This added analysis is described in section 2.2 in the revised manuscript. Briefly, temperature and relative humidity profiles were used to estimate the mixing height and cloud height over the collection campaign. Both these parameters were considered as potential influences on the deposition process: mixing height essential as a dilution factor on the ambient concentration, and cloud height as a controlling factor on the height of the atmospheric column scavenged by below-cloud wet deposition. These characteristics have been added to the revised Figure 4 and compared to the observed trends in deposition velocity. While the heights do show moderate correlation with the effective deposition velocities, their correlation is lower than that of temperature. Furthermore, the heights are observed to be primarily episodic, unlike the distinct seasonal exhibited by temperature. Hence, the hypothesis that cloud phase may be an important factor in seasonal changes in deposition

processes over the cold season was maintained. Furthermore, the radiosonde data showed temperatures above -20 °C to be common within the cloud layer in the warmer months. This supports the potential presence of mixed-phase clouds during this time.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-944/acp-2016-944-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-944, 2016.

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Discussion paper