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

Anonymous Referee #1

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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 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.

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.

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).

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.

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.

Detailed comments referenced to page/line #(s)

1/16 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

2/25 dependent on snow type

2/27-29 not sure what the message of sentence starting "The efficiencies…" is supposed to be

2/30 Last sentence is quite true, and for pretty good reasons. Why are you bucking the trend?

3/11 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.

4/16 What were the QC runs?

4/31 How, exactly, was "mass recovery precision" determined? Sounds like some kind of yield tracer was added, but no details given.

5/4-6 When was this "internal standard" added? Before or after digestion? Did it also serve as the yield tracer to track recovery?

5/11 in a class 100

5/25-26 Are U. Toronto MA thesis publicly available? If not, are they citable in ACP?

6/14-24 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.

6/24-25 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).

6/27 Flux per day over 2-18 days makes no sense for a wet deposition event likely lasting just hours. Understood that you can not separate wet and dry in this data set, but this contributes to discomfort with the calculated EVDs.

8 (Fig 1) 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).

9/13-16 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.)

9/16-18 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).

9/21-27 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.

9/28 As noted earlier, summarizing 7 months in box and whisker plot makes little sense to me.

10/10-13 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)

11/5-6 Not so sure this is true. The Hi-Vol sampling at Alert used to use cellulose filters

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Discussion paper

(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.

10/7 to 11/8 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.

12/14 enhanced deposition

12/18-21 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.

Pages 12-14 (Figs 4 and 5) 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, C204, 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.

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