

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

### **Anonymous Referee #2**

Received and published: 6 January 2017

This manuscript describes measurements of the chemical composition of freshly precipitated snow from Alert, Canada and relates those snow measurements to aerosol measurements to derive deposition velocities for various species. The manuscript reads well, and describes a unique contribution to the literature and thus is appropriate, after revision for eventual publication in ACP. The work also uses a comprehensive set of multi-species analyses (BC, IC, ICP-MS) that are a valuable expansion on prior work. However, the presentation could be improved and expanded and the link to past work in this area should be more clearly presented in the main publication.

Major points to consider:

Citations to past work are under-done in main text. An example is the work of Toom-Saunry and Barrie (2002), which described a multi-year weekly snow sampling pro-

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gram from Alert, which is quite similar to this work (with average of 4 day sampling) and should certainly be mentioned in the introduction, but is not. Work at ice core sites (e.g. Summit Greenland, Antarctica) also should be mentioned even if not at the time resolution of this study.

The analysis in this manuscript is presented mostly as a deposition velocity, the ratio of two measured quantities (e.g. Figs. 2, 4, and 5), but the individual parts of this ratio (numerator and denominator) are not discussed as fully. Because both the numerator and denominator go through annual cycles, it is useful to include discussion of both quantities. Specifically, the fall into winter aerosol increase and spring into summer transition is a time of drastic decrease in aerosol loading (e.g. March-May 2015 in Supplemental Table S8), which causes an increase in deposition velocity through equation (1). It would be useful to present graphically in the main text the cycles shown in Table S8 in reference to the snow measurements in Fig. 1. In this way, the scavenging ratio can be better understood. There is also a complementarity between high scavenging and low atmospheric aerosol abundance which can be seen in the data and should be discussed. For example, an very crude order of magnitude of the aerosol lifetime can be estimated by the ratio of the boundary layer height divided by deposition velocity. For sulfate in winter, with  $v_{dep} = 0.08$ , and crudely estimated 1 km boundary layer height, this gives 14 days, and it shortens to 5 days for the "edge seasons" (S/O/My).

Although the method for sampling is unique and useful, it causes "dry deposition" to be a bit different than it might be in true snowpack. This is briefly discussed, but should be more clear. Specifically, as air pumps through actual snowpack, it is filtered by the snow. Harder et al. (1996) indicate that filtering is a significant mechanism of "dry deposition". However, the removal (prior sampling) of old snowpack in the present study causes wind pumping / filtering to not be operative. Thus, the "dry deposition" measured here may be an under-representation of true non-precipitation-related deposition to snowpack. Also, how well particles that dry deposited to the table before snowfall are sampled by scraping off subsequent snowfall is uncertain.

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Harder, S. L., S. G. Warren, R. J. Charlson, and D. S. Covert (1996), Filtering of air through snow as a mechanism for aerosol deposition to the Antarctic ice sheet, *J. Geophys. Res.*, 101(D13), 18729–18743, doi:10.1029/96JD01174.

Another related point is that some fluxes with snow are bi-directional (e.g. snow can re-emit species such as ammonia/ammonium, nitric acid/nitrate, bromide, and mercury. In this study, old snow is removed, so emissions from snow are not measurable (and all deposition fluxes are necessarily positive). This could affect interpretation of ammonium, nitrate, and bromide, and should be mentioned for mercury even through their analysis methods did not quantify it. These processes happen in the actual snowpack, and exclusion of consideration of snowpack emissions would make the deposition fluxes measured here overestimates of the net flux for these species.

Specific Comments:

page 1, line 17: The first instance of the word "which" should be replaced by "that" to read "...deposition velocities that encompass all..."

page 3, line 22: Although loss of snow from the table before sampling is mentioned later, it also needs to be discussed here.

page 4, line 10: This reference to a thesis (which is not peer reviewed and may not be always accessible) should be replaced by a brief discussion in the supplemental material. There should not be references to theses in general.

page 6, line 9: The actual quantification of the trajectories should be described here (that is a component of methodology) instead of later.

page 6, line 17: It is also not clear if analytes deposited to a bare table would be effectively sampled from the table into the bottles, so "early dry deposition" could have a number of uncertainties.

page 6, line 19: Are these four occasions of "resuspended" snow the cause of missing data in mid winter discussed later? Make more clear if these refer to the same events.

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page 7, line 2: By "free" form, do you mean "dissociated ionic" form? It would also be useful to give some discussion of whether soluble or insoluble species dominated for various class of metals.

Table 1. At some point, method detection limits should be discussed, as well as blanks. For example, the use of nitric acid to clean bottles could lead to a high nitrate blank.

page 9, line 16: This unpublished work is not cited properly. Additionally there should be mention of findings that aerosol (particles and gases) vertical distribution are layered in the Arctic (e.g. Brock et al., *Atmos. Chem. Phys.*, 11, 2423–2453, 2011), and older "Arctic Haze" literature.

Figure 2. I'm confused about what is shown here. It appears that the minimum (presumably the actual lowest value, while prior table showed 25th percentile), median, maximum (similar question). If that is the case, I'm not sure what the "error bars" are. The error bars also seem to be added at the edges of the "box"? Overall, this looks like a "box and whiskers" plot, but it doesn't seem to have the same information as a standard box and whiskers. Please explain further.

Figure 4. In panel a, the "normalized effective deposition velocity" is shown. I'm not sure what that is. The text description doesn't really help much. Is the idea that aerosol particle components are co-deposited and that aerosol particles are internally mixed? Wouldn't I expect all aerosol particle components to then have the same deposition velocity? Figure S3 shows that different components seem to have different average deposition velocities, which could be an indication of external mixture of aerosol components, as was discussed for BC. Some further discussion, and potentially elevation of Figure S3 (or some subset of the species) to the main text would be preferable to the "lumping" that was done in Figure 4, panel a.

Figure 5. I still don't understand the error bars. Please explain.

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

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