I have read the revised version of the manuscript in detail and with great interest. I greatly appreciate the authors making the effort to address reviewer comments. I believe that the manuscript is now much strengthened and readable. Below I have listed some comments, which in my opinion still require some attention.

Specific comments

Both in the abstract and the text it is stated that the study analyzed long-term air concentrations, wet deposition, and precipitation acidity at 30 Canadian sites. The list of sites in Table 1, and Figure 6 and 8, however, include a total of up to 31 sites. Figure S1 shows the location of 29 sites only. These discrepancies are probably because of the duplicate stations in Egbert and Goose Bay, so, is 31 the final number of sites? If so please correct that number in the abstract, in the caption of Figure S1, and throughout the text.

Furthermore, differences in the availability of measurements among sites and years limited the comparison of results to a shorter number of locations (those with more than 9 years of data). As a result, the data presented in figures and tables, as well as the calculated averages, ranges, and trends mentioned and compared throughout the text, do not include results from all 31 locations in a consistent manner. For example, 16 sites were used for rates of change in annual air concentration/wet deposition data shown in tables 1, 2, and 3, while 12-14 sites were used for geographical patterns in air concentrations in figure 1, or 31 sites for wet deposition in figure 6. I imagine that for those same problems with data availability, the X axes of left and right panels in Figure 1 do not correspond to the same stations. Cree Lake and Montmorency data is shown only in 1983-1996 panels, while Bratt's Lake, Sprucedale, Frelighsburg, and Lac Edouard data appear in 1997-2010 panels only. Please summarize in the methods section the number of stations and/or time period used for each of the analyses on each of the subsections, as well as clarify the number of sites used for the calculation of averages and ranges presented in the text.

I am still not comfortable with the correlation analyses carried out between meteorological variables and particulate ions and trace gas data presented in page 10 (lines 14 to 34). I do not think that monthly averaged data correlations (or lack of) provide much information about the influences of temperature, precipitation, or relative humidity on observed K+ and NH4+ long-term trends. The use of Pearson correlation coefficients based on monthly averages to explain the long-term trends based on annual values seems a priori not very convincing. This is a complicated issue, as we are dealing with substances that differ in nature, origin, chemistry, and interactions with other pollutants. There is much evidence that current and future climatic variability and trends modulate the magnitude of annual emissions for substances like ammonia (Sutton et al. 2013). It is possible that Pearson correlation analysis of monthly data was not the best choice here. Considering that these analyses do not contribute much to the overall manuscript focus and goals, nor essential to support authors' main conclusions, I would suggest to eliminate these paragraphs in the final version of the manuscript. If necessary to support some of the statements authors made, reader can always be referred to the supplementary data. Doing so authors might improve clarity and it will also help reduce the sometimes "overwhelming" amount of data presented in the text, a concern that I expressed in my first round of comments.

Additional comments

Page 5, line 22: What do authors mean by “insufficient data”? Please clarify.

Page 7, line 10; Page 8, line 4; and elsewhere: authors refer to Fig. 1a and 1b while there are no such a and b panels, or they have not been labeled. Maybe left and right?

Figure 1: a 3-page figure seems a bit excessive. First, many of the elements should be deleted. Y-axis title (air concentration ug m-3) is the same for all panels so it can be placed just once, centered on the left. Y-axis tick labels and scale are identical between left and right panels so they do not need to be present on the right panel axis. X-axis labels are the same for the different substances so they only need to be shown on the bottom panel X-axes. Ion/trace gas labels only need to be shown in one of the two panels and not both. These adjustments will reduce the size of figure 1. Second, as a suggestion, authors might want to consider combining both panels in one single graph per substance, showing boxes of different colors (one color for 1983-1996 and another color for 1997-2010, stations that have data for one period only will show one box only).

Figure 6: As said for figure 1, please allocate X-axis labels on the bottom panel only, and just one Y-axis title centered.

References cited

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