Response to Reviewer 2
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We thank the reviewer for the suggestions to improve our paper. Our point-by-point responses to each comment are shown in blue.

This manuscript presents a detailed analysis of the trace gas concentrations and wet deposition fluxes measured at the Canadian Air and Precipitation Monitoring Network (CAPMoN) sites in the past thirty years (1983-2011). Long-term trends of aerosol ions and precipitation acidity at the Canadian sites are assessed from the aspect of acid deposition. The scavenging ratios for air pollutants as a measure of the wet scavenging efficiency are also estimated and compared with previous studies. The manuscript also presented a first attempt to quantify the relative contribution of gas and particle to measured wet deposition fluxes of ammonium, sulfate and nitrate.

Overall, the manuscript presents a very comprehensive summary of the air pollutant and wet deposition measurements at the Canadian network. While I also think that the manuscript can be difficult to follow due to the intense statistics reported in a parallel way, it is valuable to document the thirty-year dataset over Canada and the spatial and temporal patterns. The approach to estimate gas vs. aerosol contributions to wet deposition fluxes will be particularly helpful for model evaluation.

Specific comments

1) Abstract, Page 1, Line 19: Suggest change “because of the exclusion of gas scavenging” to “because of the exclusion of gas scavenging in previous studies” to avoid confusion.

Response: Revised according to your suggestion.

2) Page 5, Line 10-13: This sentence is unclear. What is the difference between the Seasonal Kendall test and the Mann-Kendall test? Why are they inconsistent? Please clarify.

Response: The Seasonal Kendall test analyzes the temporal trend in the average air concentrations in each month separately and then aggregates the results to obtain the annual trend. In the Mann-Kendall test, the data are not split into twelve months before the temporal trend is analyzed (Gilbert, 1987). Therefore, the latter was used to obtain the annual total wet deposition trend. This has been clarified in sect. 2.2.1 of the revised paper.

3) Page 5, Line 13-14 Page 10, last paragraph: “The relationship between meteorological factors and temporal trends in air concentrations and wet deposition were also examined by correlation analysis”. As shown in Table S1 and discussed on Page 10, the correlation analysis was only applied to some particulate ions and trace gases, and not to wet deposition fluxes.
It is not convincing that we can use the correlation coefficients based on monthly averages to explain the long-term trends based on annual values. For example, the weak correlation between monthly K+ concentration and precipitation may largely differ from their annual correlation, which is a better indicator for the descending trends in K+ concentrations during 1993-2010.

Please clarify. Also please state in the title of Table S1 that the correlations are computed using monthly values.

Response: The air concentrations are more likely to be influenced by meteorological parameters (e.g. temperature, relative humidity, and precipitation) than wet deposition of pollutants. Wet deposition of the pollutants is predominantly affected by the air concentrations and the precipitation amount. Thus for wet deposition, we already discussed these correlation analysis results on p. 14 lines 16-32 of the ACPD version. To clarify, we revised the sentence before sect. 2.2.2 to, "Correlation analysis was performed between monthly averaged meteorological parameters and particulate ions and trace gases. For wet deposition of inorganic ions, the correlations with the precipitation amount and air concentrations were examined." The caption for Table S1 was also revised to, "Pearson correlation coefficients between selected atmospheric ions and meteorological parameters (significant at $p<0.05$; otherwise non-significant (ns)). Note that the ion concentrations and meteorological parameters are monthly averages."

Regarding the analysis of annual atmospheric K^+ and annual precipitation amount, there was a statistically significant decreasing trend in atmospheric K^+ for almost all the sites (Table 2, see the slope and C.I.); however, no statistically significant trends in the precipitation amount were found at the majority of the sites (Table 4, see the slope and C.I.). Note that it is important to consider the C.I. (90% confidence interval of the slope) to assess whether the trend is statistically significant. The results indicate the annual atmospheric K^+ and annual precipitation trends were not the same.

4) Page 6, Line 7: Please clarify here how W(K/2) as the scavenging ratio for coastal site is calculated here. As half of W(K)?

Response: Atmospheric K^+ has a bimodal particle size distribution. It is predominantly associated with fine particles at inland locations, but also associated with coarse sea-salt aerosols at coastal locations. In our previous study (Cheng et al., 2015), we observed that the mean scavenging ratio of fine particles was 34 to 52% of that of coarse particles at inland locations, but was 80% at the coastal sites. Therefore, the fine scavenging ratio was reduced by about a factor of 2 at coastal locations to take into account that the K^+ that may be associated with coarse aerosols. This explanation has been added to sect. 2.2.3 of the revised paper.

5) Page 7, Equ. (4)(5): Replace the * with cross using the standard equation format.

Response: Equations 4 and 5 have been revised according to your suggestions.

6) Page 10, Figure 2-4: The manuscript concludes that the temporal trends in atmospheric inorganic concentrations are consistent with their emissions. Can you please also present the trends in percentage to discuss that to what extent the trends in pollutant concentrations reflect trends in emissions?

Response: The temporal trends expressed as percentages have been added to the captions of Fig. 2 to 4. In Fig. 2b between 2002 and 2010 (as discussed in the text), atmospheric ammonium at agricultural sites decreased by 6.3% while ammonia emissions in Ontario and Quebec decreased by 2.4% and 1.1%, respectively. For atmospheric nitrate and NOx emissions (Fig. 3), the timing of the trends were not synchronized as discussed in the text. In Fig. 3 between 1991 and 2001 (as discussed in the text), atmospheric nitrate increased by 3.6%. However, the period of increasing NOx emissions in Canada was between 1991 and 1997 (at 8.5%). Then, from 2001 to 2010, atmospheric nitrate decreased by 6.5%. However, the period of decreasing NOx emissions in Canada was between 1997 and 2010 (at 25.8%). In Fig. 4 between 1990 and 2010 (as discussed in the text), atmospheric sulfate decreased by 4.5% at southern Ontario and Quebec sites and by 3.6% at other sites. Over the same period, SO_2 emissions decreased by 1.8% in Canada and 4.3% in the U.S. Note that the percentage change in pollutant concentrations may not be the same as the percentage change in emissions because the quantities are very different. However, it is clear that the direction of the trends is consistent.

7) Page 34, label of Figure 3: Please explain ‘Cdn’ in the caption. And ‘Nox’ should be ‘NOx’.

Response: Cdn is the abbreviation used for Canada. We have added this explanation in the captions for Fig. 3 and 4 and changed Nox to NOx in the legend.

8) Page 42, caption of Figure 10: Suggest change here (and a few places in the text, e.g., seasonal trend) “monthly trend” to “monthly variation” to avoid confusion with the long-term trend.

Response: In the revised paper, supplementary material and Fig. 10, we replaced “seasonal trend” or “monthly trend” with “seasonal variation” or “monthly variation”.

9) Supplement, Page S5, caption of Figure S4: “Five different months” should be “five different years”.

Response: This has been corrected in the revised paper.