

Interactive comment on “Long-term air concentrations, wet deposition, and scavenging ratios of inorganic ions, HNO₃ and SO₂ and assessment of aerosol and precipitation acidity at Canadian rural locations” by Irene Cheng and Leiming Zhang

Anonymous Referee #1

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General comments The manuscript includes updated long-term data on concentrations, wet deposition, and scavenging ratios of atmospheric pollutants in Canadian sites. As outlined briefly in the introduction, there is, indeed, a need for keeping track of the recent evolution of atmospheric pollutants contributing to smog and acid rain, particularly in Canadian sites. Much of the information in the literature refers to datasets from the contiguous US or from European sites, two other regions where data from similar extensive monitoring networks have been available for decades, while Cana-

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dian sites have received relatively less attention. Observations of a recent decrease in precipitation acidity in North America (including Canada) and Europe have been accumulating, and have been the subject of many papers. The topic is not necessarily new. However, many aspects of the chemistry, transport, and deposition of these atmospheric pollutants are still not well-understood. Despite the inherent interest in reporting updated geographical patterns and temporal trends of atmospheric pollutants, much of the data presented here is not necessarily “new”, and similar reports and conclusions can be found in the literature. For example, a recent global assessment of precipitation chemistry and deposition of these substances already includes much of the information presented in the manuscript (Vet et al. 2014). The report from Vet et al. (2014) not only includes much of the data used here (data obtained from the same CAPMoN network in the same locations), but also allows to put the data into a regional context, and compare the observed geographical patterns and temporal trends with those of emissions. Surprisingly, the Vet et al. (2014) assessment was not mentioned in the manuscript. In my opinion further attention should have been paid to clarify, complement and update the information already existing in the literature. I would recommend authors further efforts summarizing and presenting measurement data and trends in a clearer geographical/temporal context. According to the four objectives presented in the introduction some of the questions that needed to be answered in the manuscript were for example: as of 2011, how much have concentrations/deposition of the several substances decreased compared to a baseline year? Where? When? Are those changes parallel to emission reductions? Much of the text is a never-ending compilation of ranges, averages, medians and percentiles of the various substances, sometimes referred as measured in site X or site Y (a non-Canadian reader must check supplementary Figure S1 constantly to figure out these locations), without a clear message being told. The reader gets usually overwhelmed by the amount of values, ranges and percentages presented in the text, much of which could just have been summarized in figures and tables, while getting very little or disperse information about the magnitude of changes that have taken place, and the spatial and temporal context of

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those changes. In my opinion, the major contribution of the manuscript is the calculation of scavenging ratios and the development of an approach to estimate particulate and gaseous wet scavenging contributions to wet deposition at those sites. Again, issues arise regarding the way scavenging data is presented and contextualized. Is there any clear spatial pattern in scavenging ratios? Have those ratios change as concentrations decreased over the decades? Have the relative contributions of particulate and gaseous substances to deposition changed over time? Specific comments Please avoid including too much data (if any) in the abstract. The message is lost among the many references to substances, ranges, years, ... In section 2.1.4. authors stated that meteorological data was collected. Apart from precipitation, were these data included in the analyses? Are there any relationships between pollutants and wind, relative humidity or temperature data? Figure 1 is a very poor attempt to show the geographical and temporal changes in concentrations. First of all, the log scale makes it difficult to note the differences among sites or the changes between the two periods. Furthermore, what's the reason behind the two periods (1983-1996 and 1997-2010)? Is 1997 a "landmark" for Canadian emission regulations that would define a "before" and "after"? Why not comparing a year in the 1980's and a year at the end of the series (2011?) to actually show the decreases? In many cases (e.g. for NO₃) the graphs show little changes or even increases, while in the text it has been clearly stated that concentrations of most substances have decreased. The case of NO₃ is particularly interesting. Figure 3 shows that averaged NO₃ concentrations and emissions started to decrease in 2001. Why not using that year to compare "before" and "after" concentrations for that substance? The multi-axis panels in Figure 5 are probably not the best option here. I would recommend to use cation/anion ratio data only for panel a (concentrations can be extracted from tables and text), and c/a ratio and pH data only for panel b (this time 2 Y axes can be used to highlight temporal trends). As mentioned in the general comments, some of the most interesting contributions of the manuscript are in my opinion those regarding scavenging ratios. Much of that information is included as supplementary materials. Some of those graphs and tables could have been part of

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the main body text. References cited Vet, R., R. S. Artz, S. Carou, M. Shaw, C. U. Ro, W. Aas, A. Baker, V. C. Bowersox, F. Dentener, C. Galy-Lacaux, A. Hou, J. J. Pienaar, R. Gillett, M. C. Forti, S. Gromov, H. Hara, T. Khodzher, N. M. Mahowald, S. Nickovic, P. S. P. Rao, and N. W. Reid. 2014. A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus. *Atmospheric Environment* 93:3–100.

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