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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The sensitivity of $PM_{2.5}$ acidity to meteorological parameters and chemical composition changes: 10-year records from six Canadian monitoring sites" *by* Ye Tao and Jennifer G. Murphy

Anonymous Referee #3

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The paper used E-AIM model to calculate the pH of six Canadian cities over the course of 10 years. The paper states that (1) summer pH is about 1 unit lower than winter aerosol pH; (2) the pH is dependent more on meteorological conditions in the summertime. In winter time both chemical composition and meteorological conditions influence the pH.

This paper is written clearly with a dataset that is suitable to be published in ACP. As the author stated, this paper is probably "the first long-term aerosol pH study in Canada" and provides the "longest records for evaluation of trends in the world". I enjoyed

reading the paper and suggest the authors consider the following before having the manuscript published.

The author made a conclusion on the effects of temperature on aerosol pH in page 6, line 20-22: "This result suggests the central role of meteorological conditions, especially temperature, in the determination of aerosol pH seasonal cycle in mid- and high-latitude regions. " Despite that the evidence raised by the author did support this argument, I think extending the 6 city dataset to all "mid- and high-latitude regions" is a bit too strong. For instance, what if there are farmland areas where ammonia emissions have a strong seasonal cycle. Then the temperature factor may not be the only dominant factor. I would advise the author to make some restrictions on this sentence.

One of the key assumptions in Figure 5 and 8(a) is that the sulfate:TNO3 ratio is 1:2. The author backed up this assumption in the text because in Toronto the average ratio of sulfate:TNO3 over the course of 10 years is about 1:2. However, since figure 5 examines the effects of chemical composition and temperature on pH from a seasonal perspective, is the sulfate:TNO3 ratio still 1:2 for each season? The author should show seasonal averaged information of sulfate:TNO3 to support that the ratio is still $\sim$1:2, otherwise a sensitivity study of how sulfate:TNO3 influences the pH should be shown in the manuscript. Figure 5 should also simulate each season based on the actual sulfate:TNO3 ratios.

The authors stated that "Noticeably, when aerosol pH shows a decreasing trend with TNO3 concentration, there is still excess NH3 in the gas phase" in page 9, line 17. Theoretically, there will always be some level of NH3 in the gas phase, despite the absolute value might be small. Therefore, the author should define what "excess NH3" means here. Maybe a plot of NH3 concentration as a function of TNO3 concentration should be shown in Figure 8 as well to show that the NH3 concentration does not change too much with increasing TNO3 values. Maybe the authors could also add a couple of sentences to clarify why a decreasing pH would still lead to an excess NH3 concentration.

Besides the typos mentioned by the previous reviewers, another typo I noticed is that the state “Vermont” is misspelled in Figure 1.

References:

Weber, R. J., et al. (2016). "High Aerosol Acidity Despite Declining Atmospheric Sulfate Concentrations over the Past 15 Years." Nature Geosci 9(4): 282-285.

Guo, H., et al. (2015). "Fine-Particle Water and Ph in the Southeastern United States." Atmos. Chem. Phys. 15(9): 5211-5228.

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