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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Synergistic enhancement of urban haze by nitrate uptake into transported hygroscopic particles in the Asian continental outflow" *by* Jihoon Seo et al.

Rodney Weber (Referee)

rodney.weber@eas.gatech.edu

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This paper assesses the composition of PM2.5 in Seoul under different transport & meteorological conditions. The separation of data collection into periods when Seoul was impacted or not by upwind transport from China, combined with local ventilation conditions over Seoul, is clever and insightful. The result is that clear differences in the aerosol composition and processes can be identified. The authors show that highest PM2.5 concentrations occur when species transported from China are present and when there is little dispersion over Seoul, as might be expected. The unique feature is they find that these periods have enhanced inorganic aerosol concentrations and

investigate a number of possible feedbacks that could explain the enhanced aerosol concentrations under these conditions, all related to aerosol liquid water levels (ALW). This includes enhanced SO2 oxidation to form sulfate and especially the uptake of HNO3 to form particle NO3-. The specific contributions of China (SO2/sulfate) vs Seoul (NOx/NH3) on these interactions is also identified. This allows a unique assessment of possible control strategies to reduce PM2.5 mass. The main issue with this paper is the lack of NH3 and HNO3 data that are required to run the thermodynamic model. The authors should more fully assess this limitation through a detailed sensitivity analysis, but my suspicion is it will not significantly change the result.

Specific Comments.

The use of acronyms made the paper, at times, difficult to follow for me. Where it is possible, it might be better to just write out the term. A list (table) defining them could also be useful. For example, in this study SIA is just sulfate, nitrate and ammonium.

Potential NO3- sampling issues? Were samples gas denuded; seems not but maybe use of Teflon filters minimizes this possible positive artifact? Since particle NO3- is a large component of this paper, and it is known to be difficult to measure using filters due to evaporation, this issue should be discussed. That is, is it possible that ammonium nitrate is significantly under-measuring in this study?

Regarding the estimated NH3 and effect on pH. A sensitivity test is warranted, as noted by another reviewer. This could include discussing epsilon(NH4+) and epsilon(NO3-). Table 2 suggests that with epsilon(NH4+) values ranging from 0.23 to 0.5, there may be some sensitivity to [NH3]. But as noted, this may not contribute to much change in pH. Furthermore given Fig 9, in some situations this may not have a large effect on predicted NO3-. The point is, the epsilon data for NH4+ and NO3- can be used to help assess the sensitivity of the predictions to uncertainties in gas phase species that were not measured; eg, one could make a graph of epsilon(NH4+) similar to Fig 9 and than show the data for a range of estimated NH3 and HNO3 around the predicted values.

Line 319, one could be more specific here, with epsilon(NO3-) near or at 1 there is a direct relation between NOx control and particle NO3-.

Sulfate is a large component of the SIA. How does sulfate play a role in this feedback mechanism (see next)?

The idea of feed back (or sometimes called co-condensation) leading to more uptake of NH3 and HNO3 by the added liquid water is not a new concept. I suggest the authors think about it some more and add a deeper discussion. It happens for any semi-volatile acidic species that when partitioned to the particle phase significantly increases the water uptake, which then raises the pH and allows more uptake. Examples include HCl/Cl- & HNO3/NO3-. Since sulfate is not semi-volatile and highly hygroscopic the semivolatile species involved that is driving this feedback process must generally have significantly higher concentrations then sulfate, or more precisely, contribute comparable or more to AWC than sulfate, otherwise the feedback does not exist. For example, in this study if sulfate was significantly larger the nitrate, would nitrate levels increase due to uptake of water? Probably not because sulfate would then control the overall AWC. One could play around with sulfate concentrations to see when this happens. The process discussed here is very similar to that discussed in Guo et al. (2017). Also, as another example, see Topping et al (2013).

Guo, H., J. Liu, K. D. Froyd, J. Roberts, P. R. Veres, P. L. Hayes, J. L. Jimenez, A. Nenes, and R. J. Weber (2017), Fine particle pH and gas-particle phase partitioning of inorganics in Pasadena, California, during the 2010 CalNex campaign, Atm. Chem. Phys., 17, 5703-5719.

Topping, D., P. Connolly, and G. McFiggans (2013), Cloud droplet number enhanced by co-condensation of organic vapours, Nature Geoscience, 6, 443-446.

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