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Atmospheric Chemistry and Physics
Discussions

Interactive comment on “The Role of Coarse Aerosol Particles as a Sink of HNO_3 in Wintertime Pollution Events in the Salt Lake Valley” *by* Amy Hrdina et al.

Anonymous Referee #2

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Hrdina et al. present an interesting, although circumstantial, case for the importance of coarse particles as a sink of HNO3 in the wintertime in the Salt Lake Valley (SLV). The authors do not have direct measurements of coarse particle composition, but make use of PM2.5 composition measurements, coarse particle size distributions, and snowpack chemistry to make a case for reactive uptake of HNO3 on coarse particles influencing the budget of HNO3 in the SLV and associated implications for fine particle chemistry. The manuscript is overall well written and most of the analyses presented sound.

Some of the authors’ planned measurements have missing data (e.g., the PM2.5 anions during the main PM episode studied). This, along with the absence of any coarse

particle PM composition measurements, however, makes it difficult to constrain the problem well. This is compounded by issues with how snowpack chemistry is used (incorrectly, I believe) to quantify the amount of total nitrate in the atmosphere and the impacts of coarse particle reactive uptake of HNO3 on submicron NH4NO3 formation.

My specific comments follow:

1. p.2, line 27: I think it is fairer to say that automotive and industrial processes have increasingly been recognized as important ammonia sources in urban areas.

2. p. 5, line 31: the authors need to justify their choice of a density of 1.0 g/cm3 for coarse particles in calculating mass concentrations from APS measurements. This is quite low for typical coarse particle types, including the sea salt and dust considered in this manuscript.

3. p. 8, lines 16-18: the authors should provide information about the types of road de-icers used in the area surrounding their measurement site and whether they change as a function of forecast temperature. Many liquid de-icers commonly used in parts of the Rocky Mountain west include Ca or Mg. Are these used in Salt Lake City, on the UU campus? Do they change between conventional salt and liquid de-icers depending on temperature?

4. The analysis of reactive uptake lifetimes for HNO3 on p. 10 is quite interesting. The authors should extend this analysis to consider the relative HNO3 sink rates for NH4NO3 formation vs. reactive coarse particle uptake.

5. p. 11, lines 8-17: The authors here focus discussion on prior work concerning coarse particle uptake of HNO3 in coastal environments. This is interesting and relevant, but they should also cite observations of uptake in more continental environments which might be better models for the SLV. Lee et al. (doi:10.1016/j.atmosenv.2007.05.016), for example, examine the importance of coarse particle nitrate at both interior and coastal U.S. environments.

6. I have serious reservations about how the authors use snowpack chemistry observations to constrain atmospheric levels of total nitrate. As discussed in the manuscript, falling snow composition reflects some combination of in-cloud and below-cloud scavenging processes. The composition of snowpack on the ground is further affected, over time, by accumulated dry deposition. If one hypothesizes that significant nitrate is present in reacted coarse dust or salt particles, this coarse particle nitrate can be effectively scavenged by falling snow (coarse particle scavenging efficiencies are much high than scavenging efficiencies for accumulation mode particles) and is also very effectively deposited to snowpack on the ground by dry deposition via sedimentation of these large particles. NH4+ particles, on the other hand, are submicron and thus have both lower scavenging efficiencies by falling snow crystals and much lower dry deposition rates to the surface than coarse nitrate particles. What this means is that a comparison of snowpack NO3-/NH4+ ratios is not at all representative of atmospheric concentration ratios of total nitrate/ammonium. The snowpack ratio, in the presence of coarse mode nitrate, will be significantly higher than is found in the ambient atmosphere. For this reason, the author's use of this snowpack ratio as a surrogate for what was in the atmosphere is incorrect and certainly biased high. Using this ratio to estimate the impact that eliminating coarse nitrate would have on PM2.5 NH4NO3 formation will, therefore, lead to a significant overestimate. I honestly don't see how the authors can get around this limitation on the utility of the snowpack composition data. I think the fact that the NO3-/NH4+ ratio is elevated in the snowpack does help the authors make the case that coarse particle nitrate is present, but I do not see how they can properly extend the comparison to argue what the total nitrate/ammonia ratio is in the ambient atmosphere.

7. Fig. 1: Suggest changing NH3(g) and associated flux arrows to another color. The orange color used could be misinterpreted by the reader as related to daytime pathways per the description in the caption.

8. Panels (a) and (b) of Figure 2. I personally found it somewhat unhelpful to see the

PM concentrations presented in mass units while the gas concentrations are given as mixing ratios. This makes it hard, for example, to compare relative amounts of gaseous and particulate NHx in panel (a).

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