

## ***Interactive comment on “The Role of Coarse Aerosol Particles as a Sink of HNO<sub>3</sub> in Wintertime Pollution Events in the Salt Lake Valley” by Amy Hrdina et al.***

### **Anonymous Referee #1**

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This paper analyzes data from pollution events in Salt Lake City during cold periods when strong inversions lead to generation of PM<sub>2.5</sub> ammonium nitrate. The analysis is limited in that there was no overlap in all instruments during periods of interest. For example, the APS measuring coarse mode size distributions was only operational during a short period of the overall study. It did coincide with a PM<sub>2.5</sub> event, but during that event there was no PM<sub>2.5</sub> aerosol data. This makes much of the analysis more speculative. Overall, the analysis is somewhat obtuse, especially the thermodynamic analysis, which could use more explanation and additional details. It would benefit from a more rigorous approach in which particle pH and partitioning of ammonium nitrate is discussed. There are some other clarifications needed, which are discussed below.

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## Specific Comments.

Is the measurement site representative of the greater SLC area? Does its elevation influence this?

Pg 2, Lines 15 to 16 regarding the health effects of  $\text{NH}_4\text{NO}_3$ . The logic is  $\text{NH}_4\text{NO}_3$  drives  $\text{PM}_{2.5}$  mass,  $\text{PM}_{2.5}$  mass is associated with adverse health and is regulated, so  $\text{NH}_4\text{NO}_3$  has adverse health implications and should be reduced to meet  $\text{PM}_{2.5}$  regulations. This is standard logic and so a reasonable statement, but if the authors really want to be precise, there is debate if  $\text{NH}_4\text{NO}_3$  is toxic. The authors could easily add more details since health studies have been performed for the regions they are measuring, such as [Watterson et al., 2007]. The results are not as clear as the statement in this paper. Also, why not discuss an environmental effect that can be directly linked to  $\text{NH}_4\text{NO}_3$ , the effect on visibility and haze due to its hygroscopicity, and what about nitrogen deposition; the latter seems especially important since this paper is really about deposition?

Pg 5 line 3, could not find the Markovic ref. Is the chemistry of the water for the wet denuder and aerosol collector altered to help adsorb species? This is most likely an issue for the wet denuder that is collecting both anions and cations gases. That is, is an absorbing species added to liquid or is it pure water?

What is the RH of the aerosol sampled by the TEOM and APS? Are the particles dry?

Fig 1 is a nice schematic.

It might be more insightful to plot the gases in units of  $\mu\text{g}/\text{m}^3$  to allow direct comparison with aerosol concentrations, instead of mixing ratios. Although mixing ratios are more traditional.

It should be noted in Fig 3 caption or the text that the total fine mode surface area spans sizes 0.54 to 2.5  $\mu\text{m}$  since it was measured by the APS. Is that really total fine particle surface area? Are the particles dry?

Pg 9, the equation for uptake of  $\text{HNO}_3$  by coarse mode particles seems incorrect. It looks to be the formula for free molecular regime uptake, which does not apply to coarse mode particles. A correction factor due to diffusional resistance, ie something that can be derived from the size distribution and some transition regime mass transfer formula, like Fuchs Sutugin, see Seinfeld and Pandis, may be needed.

Pg 13, line 8-9, This result is expected since... Please clarify. It might be better to compare the predicted gas and particle partitioning, not  $\text{NH}_4\text{NO}_3$  predicted, which is the form that is being assumed that the model is predicting (ie, the model does not output the  $\text{NH}_4\text{NO}_3$  concentration it outputs  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , along with other species, so I assume the authors are simply taking the output  $\text{NO}_3^-$  adding the  $\text{NH}_4^+$  and comparing to a similar addition of data in the form of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ ). Aerosol pH seems to largely ignored in this work; pH values are never give despite it controlling the concentration of  $\text{NO}_3^-$ . It would be curious, for example, to know what the difference in pH is for with and without cation inclusion. This, and a focus on  $\text{NO}_3^-$  predicted vs observed would help interpret the results and provide more detail than the statement in the paper (line 8-9). It seems that what is being implied here is the model is simply doing an ion balance with the input anion and cations; ie if you add more cations than there will be less predicted  $\text{NH}_4\text{NO}_3$  since there is some  $\text{Ca}_2(\text{NO}_3)_2$  (as an example). But isn't it much more complex since adding cations changes the pH which affects the partitioning of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , (and other semivolatile ionorganic species, such as  $\text{Cl}^-$ ... ). At the higher concentrations the simulated and measured agree. Why is this happening? Is it consistent with the interpretation of what is happening at lower concentrations? The authors might also consider the assumption of mixing state, they implicitly assuming that everything is internally mixed.

The last part of the main text is also not clear (Pg 13, lines 11 to end of section). As I understand it, the idea here is that if all the  $\text{NO}_3^-$  and  $\text{NH}_4^+$  found in snow (which got there by reacting with large particles that then fall out) was instead in the gas phase or in fine particles, and  $\text{PM}_{2.5}$  nonvolatile cations were present, (but no coarse mode),

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then the predicted  $\text{NH}_4\text{NO}_3$  would be much higher (2x). But it is not clear what this proves. The situation is much more complex, and really requires a full model. Eg, would not the deposition by gases and fine mode particles change between these two cases. This is not considered. The deposition of the gas species of  $\text{NH}_3$  and  $\text{HNO}_3$  is  $\times 10$  higher than a fine particle, in the case of no coarse mode, the gas concentrations could be much higher and so gas deposition much higher, which could limit  $\text{PM}_{2.5}$   $\text{NH}_4^+$  and  $\text{NO}_3^-$  formation. This is not considered in the analysis presented. The general conclusion that the coarse mode is a sink for Total  $\text{NO}_3$  is clear, but how that impacts  $\text{PM}_{2.5}$   $\text{NH}_4\text{NO}_3$  concentrations (it may be better to just talk about  $\text{NO}_3^-$  concentrations), is not clear from this simplistic analyses.

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