Response to Reviewer #4 comments:

The authors conducted thermodynamic modeling analysis on the acidity and inorganic aerosol gas-particle partitioning in the South Korea during an aircraft field campaign. The authors also determine the "chemical regime" of PM sensitivity to ammonia and nitrate availability and found that the aerosol formation is always sensitive to HNO_3 levels, especially in highly polluted regions, while it is only exclusively sensitive to NH_3 in some rural/remote regions. This manuscript is well written and organized, and the topic of PM control strategy is relevant to this journal. However, I am concerned about the validity of the new approach.

Answer: *We thank the reviewer for the enthusiastic response and for feedback that has improved the manuscript. Below, we include the response to comments and questions raised.*

In the NPC-pH method, the authors estimated pH without using observed gaseous-NH_3 concentrations. Given that there are many measurement data of gaseous NH_3, the validity of the NPC-pH method should be evaluated by using the observed NH_3 data (e.g., Calnex or SOAS campaign in Table 1) before the application to this method to the data without the observational NH_3 data. Without this verification, the discussion on the PM control diagram with this method is less convincing.

Answer: *We thank the reviewer for raising this point. It is understandable that the reviewer recommends to evaluate the method with observational data. This is possible, but unnecessary in our opinion as the thermodynamic model has already shown to reproduce NH_4-NH_3 partitioning in relevant datasets (e.g., Guo et al., 2015; Guo et al., 2018). The thermodynamic model then can be used as a good proxy of ambient aerosol partitioning. That the algorithm works even with "noisy" data, proves that the pH estimates are robust.*

I strongly recommend the authors to conduct verification of this method from comparison of the model simulations with and without observational NH_3 data. In addition, the authors showed in Figure 3 that the simulation results diverged with the Guo method with the increase of iteration number. Here, the similar analysis with the NPC-pH method could clearly demonstrate the superiority of the NPC-pH method, so I recommend the authors to include such analysis.

Answer: *This is a good point. We will include a comparison with the Guo method in the revised manuscript.*

Specific comments:

L246-266: It is difficult to understand the methodology and the results of this analysis. How did you combine datasets of total NO_3, total SO_4, and total NH_4? How many datasets did you prepare? Why did the authors evaluate the NPC-pH method by inputting total NH_4 into the model (and not NH_4^+ as in Figure 4). In which condition (e.g., ranges of inorganic component concentrations), the results of NPC-pH method deviate from those of equilibrium method (Figure 5).

Answer: *The datasets are assembled from the observations of gas-phase HNO_3, aerosol NO_3 (which summed together gives total NO_3), aerosol sulfate (which is the total SO_4), and aerosol ammonium (which is set to the initial total NH_4 in the algorithm). The NPC-pH method tends to*

be subject to some noise when the aerosol nitrate and aerosol pH are both low. We clarify these points in the revised text.

Figure 1: I'm afraid that the data in this figure is not strictly PM1, but include PM with a size < 4.1 micro-m (NVS).

Answer: *Thank you for pointing this out. We have added these clarifications in the revised text.*

Table S1 (L374-375): I recommend the authors to add average temperature and RH, so that analysis of Figure 9 could be more informative.

Answer: *Good point! The data will now be included in the revised supplement*

References

Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite Jr., J. R., Carlton, A. G., Lee, S.-H., Bergin, M. H., Ng, N. L., Nenes, A., and Weber, R. J.: Fine-particle water and pH in the southeastern United States, Atmos. Chem. Phys., 15, 5211–5228, https://doi.org/10.5194/acp-15-5211-2015, 2015.

Guo, H., Otjes, R., Schlag, P., Kiendler-Scharr, A., Nenes, A., and Weber, R. J.: Effectiveness of ammonia reduction on control of fine particle nitrate, Atmos. Chem. Phys., 18, 12241–12256, https://doi.org/10.5194/acp-18-12241-2018, 2018.