Response to Reviewer #2 comments:

The manuscript "Aerosol pH and liquid water content determine when particulate matter is sensitive to ammonia and nitrate availability" by Nenes and co-workers presents … a regime where neither NH_3 nor HNO_3 is important for PM levels. This latter regime is perhaps the most interesting aspect of the paper as, to my knowledge, it is the first time it has been pointed out. The manuscript presents results that are of interest to the scientific community, it is well written and overall the concepts are expressed with clarity.

We want to thank the reviewer for the positive and thoughtful comments that improve the manuscript. Below, we include the response to comments and questions raised and outline the changes made to the text.

Reviewer comment: “The main criticism I have is that while the authors explain well when this framework works well, they don’t clarify when it is necessary to pay extra care at using it e.g., temperature below 273 K? Low aerosol water content? And in what instances aerosol pH and/or water content calculations are less reliable (as those quantities cannot be measured directly)”.

Answer: *These are all important points. The framework works well, as long as the assumption of thermodynamic equilibrium and a single aqueous phase provides a good representation of the aerosol. Typically this is associated with humidity above 40% and for temperatures about which mass transfer limitations are not severely limited by highly viscous or semi-solid aerosol. This is discussed extensively in Pye et al. (2019), Battaglia et al (2019) and others.*

Reviewer comment: “Additionally, a mention on how the presence of an organic fraction in the aerosol mass could affect the pH and aerosol water content would help to guide the reader towards better use of this framework. I think that adding a sentence or two addressing the possible pitfalls that could occur in using this framework in the wrong domain would be of great help to the readers”.

Answer: *Thank you for pointing this out. As formulated here, the framework does not imply that the water is associated with the species considered (ammonium, nitrate) but rather it is treated as a variable; pH is also treated as a variable and can be modulated from organics, NVCs, halogen ions, sulfates, carbonates, and other species. The main requirement is that the aerosol is dominated by a single aqueous phase, as discussed in Battaglia et al. (2019).*

Reviewer comment: “page 1 line 23: the four regimes are named in a different way here than in the main text. The naming used in the abstract is somehow confusing as "NH3-dominated" and "HNO3-dominated" does not immediately tell if the regime name refers to the aerosol phase, gas phase or total (aerosol+gas) therefore the reader could have a hard time to understand if the "NH3-

dominated" regime is the same or the opposite than the "NH3 sensitive" regime. I recommend harmonizing the name of the four regimes and adopt in the abstract the same clearer nomenclature used at page7 lines 9 to 15 in the main text".

Answer: *Thank you for noting this. We have addressed this issue now.*

Reviewer comment: "page 1 line 24: "... neither NH3 and HNO3 ..." maybe "... neither ... nor "?".

Answer: *Done*

Reviewer comment: "page 6 line 3: "... the e(NO3-) is nearly 1 and almost all nitrate (NO3_T) is in the gas phase (HNO3)" This sentence is confusing. If the fraction of total nitrate in the aerosol phase is near 1 how can it be that almost all nitrate (NO3_T) is in the gas phase?".

Answer: *Thank you or pointing this out. It was a typo, which is subsequently addressed.*

Reviewer comment: "Table 1: indicating the units in the header (not only in the caption) would be useful to the reader".

Answer: *Done*

Reviewer comment: "Figure 5: indicating the temperature ranges for each data set would be useful to the reader".

Answer: *We have noted in the caption that the average temperature, humidity and composition – along with their variances – are provided in Table 1.*

Reference:

Battaglia Jr., M. A., Weber, R. J., Nenes, A., and Hennigan, C. J.: Effects of water-soluble organic carbon on aerosol pH, Atmos. Chem. Phys., 19, 14607–14620, https://doi.org/10.5194/acp-19-14607-2019, 2019

Pye, H. O. T., Nenes, A., Alexander, B., Ault, A. P., Barth, M. C., Clegg, S. L., Collett Jr., J. L., Fahey, K. M., Hennigan, C. J., Herrmann, H., Kanakidou, M., Kelly, J. T., Ku, I.-T., McNeill, V. F., Riemer, N., Schaefer, T., Shi, G., Tilgner, A., Walker, J. T., Wang, T., Weber, R., Xing, J., Zaveri, R. A., and Zuend, A.: The Acidity of Atmospheric Particles and Clouds, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-889, in review, 2019