Response to Reviewer #1 comments:

This is an excellent and important paper that introduces a new conceptual model for different aerosol "chemical regimes" based on a simple framework formalized around ammonia and nitric acid partitioning between the gas and aerosol phases as a function of aerosol pH and liquid water. The paper is well written and easy to follow, and provides a very useful way of quickly identifying the aerosol chemical regime, and more specifically, the sensitivity of particulate mass to changes in gas phase ammonia and nitric acid emissions. This new framework will be extremely useful for efficiently employing models to evaluate expected changes to particle mass loadings given different emissions scenarios, which will be useful for informing policy makers.

We thank the reviewer for the enthusiastic response and thoughtful comments that have improved the manuscript. Below, we include the response to comments and questions raised.

Reviewer comment: "One question I have is whether there might be other potential variables of importance (in addition to temperature, which the authors of course acknowledge)? For example, ionic strength, especially at very low LWC values."

Answer: *The reviewer raises a good point. Ionic strength is indeed a variable that affects the analysis and resulting maps – and is already considered by the framework in the Φ, Ψ terms. Given that the pH vs. W curves depend logarithmically on Φ, Ψ (hence activity coefficients), ionic strength impacts on the pH is in general of second order. For this reason, we consider the dataset-average activity coefficient when plotting the relevant curves.*

Reviewer comment: "I also wonder, when evaluating real ambient aerosols, how organic compounds could skew these results, especially since organics can comprise a substantial fraction of the aerosol mass (and therefore potentially impact both aerosol pH and liquid water). The authors do not discuss organic aerosol at all, which seems to be a big oversight. However, this closing sentence makes it clear that the authors are aware of potential biases like this, and that these potential biases do not detract from the basic goal of the framework, which includes identifying ": : :regions or time periods where pH and liquid water content prediction errors matter for PM sensitivity assessments.""

Answer: *This is an excellent point, and we should have discussed this more in the original manuscript. The effect of organic species – in terms of their impacts on liquid water and activity coefficients – is secondary, as long as the aerosol is a single phase (Battaglia et al., 2019). As long as the single phase requirement is satisfied, the framework will provide quite plausible results.*

Reviewer comment: "While the different regimes are very useful in their own right, I was expecting that the lambda and kappa values described in the methods section would be used later in the paper to further characterize the aerosol mass sensitivity (e.g. not just *that* the aerosol is sensitive to NH4 emissions, but *how* sensitive). This might be a nice addition, to further show how the data could be used by policy makers."

Answer: *This is a very good suggestion. We have added such points in the revised text.*

Reviewer comment: "'NVC' used but not defined in the abstract."

Answer: *Thank you for pointing this out. It is now addressed.*

Reviewer comment: "Why use "kappa" for the ratio of PM mass formed per mole of NO3, when kappa is already well-known and widely used by the aerosol community as the hygroscopicity parameter? Is there no other Greek letter available?".

Answer: *Indeed so. We have replaced "kappa" with the Greek letter "zeta" (ζ).*

Reviewer comment: "The examples given for lambda are helpful, but more extreme examples are also possible (e.g. ammonium bioxalate, lambda = 142/14 = 10)".

Answer: *The discussion in the original manuscript focused on the most likely values, given what we know about aerosol composition. However, we will note this extreme value, even if $\lambda=10$ assumes that oxalic acid resides completely in the gas phase before it condenses to the aerosol in the form of ammonium bioxalate; pure oxalic acid can reside substantially in the aerosol (~50%) regardless of pH (Nah et al., 2018), therefore λ can be considerably less than 10.*

Reviewer comment: "Typo in table 1 caption: (ug u-3)".

Answer: *Corrected.*

Reviewer comment: "In Fig. 4 it is unclear why the shaded regions extend to epsilon of 0.2 (rather than 0.1)".

Answer: *Thank you for pointing this out. The issue is now corrected.*

Reviewer comment: “Line 26 on Page 9: “(keep or delete)” I suppose you should keep this statement (“NVCs from sea-salt”). Of course, the mixing state of the NVCs is central to their impact. This should probably be stated, succinctly, if the statement is indeed kept”.

Answer: *Thank you for these suggestions. The manuscript has been modified as suggested.*

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

Nah,T., Guo,H., Sullivan, A.P., Chen, Y., Tanner, D. J., Nenes, A., Russell, A., Ng, N. L., Huey, L. G. and R. J. Weber: Characterization of Aerosol Composition, Aerosol Acidity and Organic Acid Partitioning at an Agriculture-Intensive Rural Southeastern U.S. Site, Atmos.Chem.Phys. 18, 11471-11491, https://doi.org/10.5194/acp-18-11471-2018, 2018