

Interactive comment on “Aerosol pH and liquid water content determine when particulate matter is sensitive to ammonia and nitrate availability” by Athanasios Nenes et al.

Anonymous Referee #1

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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.

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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.

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."

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.

————— Minor comments: 'NVC' used but not defined in the abstract.

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?

This statement is a bit open-ended: "...if nitric acid were replacing chloride in seasalt (e.g., conversion of NaCl to NaNO₃), the ratio would be $\kappa = (85-58.4)/62=0.43$ " This seems to imply that the Cl (from NaCl) and H (from HNO₃) evaporate from the aerosol as HCl. If that is the assumption, this should be stated explicitly. If volatilization of CO₂ is assumed to occur during reaction between alkaline dust and HNO₃, this should also be stated explicitly, for clarity.

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

Typo in table 1 caption: (ug u-3)

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

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.

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