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Comment on acp-2022-815

Anonymous Referee #1

Referee comment on "Effects of simulated secondary organic aerosol water on PM_1 levels and composition over the US" by Stylianos Kakavas et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-815-RC1, 2023

Review of "Effects of Secondary organic aerosol on fine PM levels and composition over the U.S." by Kakavas et al.

This manuscript describes air quality model simulations with PMCAMx of North America with focus on the U.S. The model simulations include ISORROPIA-lite to simulate aerosol liquid water not only from inorganic PM constituents, but with water uptake contributed from secondary organic aerosol (SOA) constituents. The authors investigate the amount and relative change in aerosol water, dry mass from nitrate, HCl, HNO3, and ammonium due to this process. The authors find ubiquitous increase in predicted wet and dry PM1 mass concentrations. The authors identify an important and interesting topic regarding interactions among water uptake, organic aerosol species formed in situ and the impacts on particle-phase chemical composition.

There is no connection of model predictions to measurements and it is difficult to understand if the changes represent improved predictive skill. The authors generally provide little to no support for employed values (e.g., kappa, density) or the city selection. If statistical tests were performed, for example, to determine that the 1% difference in PM1 dry mass is statistically significant – they are not discussed. Would such change be sufficient to be detected in an observational network or during a field campaign?

I cannot support publication of this manuscript in its present form. Provided the comments below are addressed the manuscript may be publishable.

Detailed Comments:

This a 3-D modeling study, and authors make no connection to field observations. The title should reflect that. For example, "Effect of simulated …."

The authors motivate their work with discussion of PM2.5, and classify all of their results in terms of PM1. Why the disconnect? Further, how was [PM1] calculated from model output? This is not described in main text or supplemental information.

Line 37: "Potassium levels can be significant … biomass burning" This sentence seems a little out of place, especially given the Cl discussion regarding biomass burning later in the manuscript (lines 171-173). Also, is the review paper by Pye et al, the best reference for this point?

The HCl hotspot in KS should be addressed. In the text, chlorine species are discussed primarily in relation to their presence due to biomass burning, which (I don't think) is happening in the KS hotspot.

Line 50: Does "a lot more hygroscopic" have a quantitative meaning?

Line 117 and Line 119: can justification be provided for the Kappa and SOA density values?

Can the authors defend the use of the kappa values in the context of a regional simulation or evaluation focused on discussion of urban areas? In the simulations here, aromatic SOA has the same hygroscopic properties and density as 'aged' SOA? Why not pick a higher and lower bounds-0.3 to 0.05? Or better yet, why not apply k values based on chemical information of SOA species since the authors have that information from the model? Any which way, some reasoning for the chosen kappa values is needed.

Why were the particular cities selected? Why are they introduced in the end?

It is difficult to accurately measure RH above ~95%. Did the authors screen out any RH values when evaluating water mass predictions?

The authors state "The model performance has been evaluated for fine PM and its components for the examined period by Skyllakou et al. (2021)." What did they find? For example, in these simulations there is a universal increase in PM1 mass. Was such a one-way bias observed in Skyllakou? Does this model configuration address model bias in a way that enhances predictive skill? From my quick read of Skyllakou it appears there is often a positive bias (overpredition) of PM2.5 mass concentrations. To what degree does this new model process exacerbate bias and error?

Starting at line 217: “Aerosol liquid water directly affects the PM sensitivity and dry deposition rates, with direct implications for emissions control policy.” It reads awkwardly to introduce these new ideas in the last paragraph of the manuscript.

The finding that increasing the amount of liquid water increasing nitrate concentrations is an important finding in the spirt and context of this sentence – but the authors gloss over this.

Table S1: Can the authors provide quantitative meaning or context for “low”, “high” and “modest”? How does RH change in these areas?

Sacramento is listed as “low” SOA in Table S1. Sacramento is one of the top 20 most polluted cities in U.S. AMS studies in Davis, CA & Cool, CA (i.e., near Sacramento) are heavily organic dominated. Can the authors defend the choice to characterize Sacramento as ‘low’?

Editorial:

The months used for the seasonal definitions are not provided.

The y-axis in the first row of Fig. S7 is log scale. Why? There should be a note in the Figure caption each time the axes differ.

The authors rely on some supplemental figures heavily, referring to them many times. They should probably be in the main text.