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Comment on acp-2021-836

Anonymous Referee #2

Referee comment on "A predictive viscosity model for aqueous electrolytes and mixed organic–inorganic aerosol phases" by Joseph Lilek and Andreas Zuend, Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-836-RC2>, 2021

This manuscript describes an exciting extension of the AIOMFAC-VISC model. AIOMFAC and AIOMFAC-VISC are models that are extensively used in the aerosol community. Until now, AIOMFAC-VISC could not perform viscosity prediction for systems other than binary organic/water. Since atmospheric aerosol particles are mixtures of organic and inorganic compounds, extending AIOMFAC-VISC to include more complex mixtures will be very beneficial.

General comments:

The new extension presented here is shown to work exceptionally well for both water/electrolyte and water/electrolyte/organic systems. The manuscript is accessible, even without a background in thermodynamic modeling. The authors certainly put in the effort to explain the justifications. I have no major concerns; the agreement between measurements and AIOMFAC-VISC are suggestive of the broad applicability, authors are forthright regarding the limitations of the model as well as the fact that there are a limited number of data sets to compare model predictions to. I recommend publication of this manuscript, which will be a tremendous resource to the aerosol community.

Anonymous Referee 1 raises an excellent point about "unphysical" predictions regarding crystallization and gelation. However, I will note that this aspect of the model may actually be particularly helpful at identifying gel/ efflorescence transitions in measurements based

on deviations from the model. For example, MgSO₄ is known to form a gel at water activity of ~ 0.3 . At this water activity, AIOMFAC-VISC predicts a viscosity of $\sim 10^4$ Pa s (from Fig. 5 of the manuscript). In Richards et al. (2020a), MgSO₄ was observed to be entirely rigid at a water activity of ~ 0.3 , which is not consistent with a viscosity of 10^4 Pa s. This discrepancy further supports the notion of a gel transition. Similarly, the discrepancies between experiment and model in Figure 8 of the manuscript, where viscosity measurements of Mg(NO₃)₂ and Ca(NO₃)₂ were compared to AIOMFAC-VISC predictions, reinforce the supposition that efflorescence, or some form of phase transition, had occurred in measurements.

I do not believe this to necessarily be a problem with the model. Rather, it is something the community should be aware of when interpreting model predictions. Efflorescence is difficult to predict without observations, and even then can occur across a range of RH (in principle, as high as the deliquescence RH). Similarly, not enough is known about gel-forming systems at this point to identify any gelation point where the model becomes unphysical. Richards et al. (2020b) has some limited observations involving CaCl₂ and organics. Nadler (2018) also suggests a gel transition in CaCl₂-sucrose systems at ~ 26 - 28% RH. Lv et al. (2018) showed gelation in magnesium acetate at $< 50\%$ RH. In other words, the available observations are quite sparse. Unless/ until there are more measurements on gel transitions/ efflorescence of all potential mixtures, the presented version of AIOMFAC-VISC is possibly the most helpful.

Minor comments on formatting, presentation, clarity, etc.

In Figures 4- 7, is there any particular reason all of the y-axes extend to 10^8 Pa s? For some systems, e.g., Fig. 7a (KBr), which has a maximum value of ~ 1 Pa s, it becomes difficult to see any differences because the values appear effectively flat relative to the large scale.

My other comments on presentation/ clarity have already been covered by Referee 1, so I will not reiterate those here.

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