This is the review for the manuscript entitled “A predictive viscosity model for aqueous electrolytes and mixed organic–inorganic aerosol phases” by Lilek and Zuend.

This manuscript describes an extension of the thermodynamics-based group-contribution model AIOMFAC-VISC to predict viscosity for aqueous electrolyte solutions and aqueous organic–inorganic mixtures. Viscosity is described by implementation of the activation energy for viscous flow following Eyring’s equations for absolute rate theory. The viscosity information of close to 30 electrolytes (inorganic salts) were fitted simultaneously with AIOMFAC ion-specific expressions. Three different approaches were examined to describe viscosity of organic–inorganic mixtures. Lastly, an example is given how AIOMFAC-VISC can be applied to predict viscosity for phase separated organic-inorganic particles.

The manuscript is well written, and its topic fits the research areas covered by Atmospheric Chemistry and Physics. I do not have major comments or revision requests, only minor comments the authors might consider before publishing the manuscript.

In general, it might be beneficial to better integrate discussion of data shown in supplement into the main text. Some supplemental data show, e.g., that it is not always clear if AIOMFAC-VISC performs better than the Laliberte model and the reasons could be pointed out in main text. Additional viscosity data for organic-inorganic mixtures in supplement appear to not be predicted as well by AIOMFAC-VISC. Gel formation, phase transitions are mentioned as a cause but not within the place of the discussion of these data.

Reading some of the sections, it occurred to me if there is a relation to the Hofmeister series (kosmotropes/chaotropes)? Just a comment.

l. 175: When reading this equation, the first question which came up in my mind is about the impact of hydration shell, shielding of ions, etc. This is addressed in the following section but how exactly was not clear to me. How does the hydrated volume impact the viscosity parameters/prediction? At that point in text, the mathematical link was not entirely clear. Also, around l. 175, one may want to make a note that hydration effects will be discussed in the next section.

l. 511-513: Could “such as that for NH4Cl shown in Fig. 2d, the model is capable of
reproducing the local minimum in measured viscosity.” be shown in an extra figure in supplement?

l. 532: I am not sure if I see that AIOMFAC-VISC outperforms the Laliberté model for 5 out of 6 points in Fig. 3b. For a mass fraction of water > 0.8, the models seem to perform similar. Below that amount of water, both models disagree with measurements (3 points) where AOIMFAC-VISC tends to be closer to the experimental data.

l. 536 and following: More discussion should be given towards data in supplement. “AIOMFAC-VISC performs as well or better than the Laliberté model” does not mean that experimental data is reproduced. Likely, a more detailed discussion is needed.

For example:

Fig. S1b, at low mass fraction of water, AIOMFAC-VISC yields higher viscosity.

Fig. S1c, d: Is something missing in legend? Symbols and legend do not fit.

Fig. S3a: Is AIOMFAC-VISC doing consistently worse?

l. 644-645: Results shown Figs. 6 and 7 are very interesting and should be more discussed in main text. The reasons for the discrepancy between 1:1 mixtures and predictions could be a bit more elaborated on.

l. 679 and following: Discussion of Fig. 12. Data for phase states of SOA and SOA with sulfates as a function of temperature and humidity are scarce. Suggestion: data shown in Fig. 12 could be set in context with the experimental/theoretical study by (Charnawskas et al., 2017) who looked at water uptake and freezing of SOA, SOA+sulfates with respect to glass transition and full deliquescence relative humidity.

Technical corrections:

l. 442: “is the data...” instead of “in the data”?

References