

Chen et al. present a new framework to predict the efflorescence relative humidity (ERH), i.e., the RH at which the inorganic material undergoes efflorescence/crystallization, in internally mixed organic-inorganic aerosol particles. The topic is interesting and fits within the scope of the journal. The manuscript is interesting, but in its current form warrants clarification and improvement in several aspects listed below, before it can be considered for publication.

**General:**

- The topic of the presented manuscript warrants a more detailed discussion of aerosol phase state and phase transitions in the introduction section.
  - o While the phase state of atmospheric organic aerosols is often described by dynamic viscosity as either liquid, amorphous solid or amorphous solid (“glassy”), some organics can also exist as crystalline solid, i.e., undergo crystallization/efflorescence. It is important to clarify early, that the ERH model presented here, is concerned about the efflorescence of the inorganic component in internally mixed organic-inorganic aerosols. An extended discussion of L159-163 should be put in the introduction.
  - o Efflorescence or crystallization of the inorganic component is a nucleation process that requires the particle to be supersaturated with respect to that inorganic material. While the authors note correctly that efflorescence is a kinetically controlled process (L245), a more detailed description of the efflorescence process is needed. Given it is a nucleation process, there is some statistical component associated with the crystallization of the salt in an inorganic-organic mixtures. Thus, if you were to take a particle of a given size and composition and make it undergo efflorescence by decreasing the humidity of the system, you would get a mean ERH value along with standard deviation, as crystallization is not expected to always occur at the exact same RH. How does your model take this into account?
- More information on the ERH-viscosity model (eqs. 4 and 5) is required.
  - o Please provide a more detailed description of the literature data of ERH and viscosity and how the model was derived from this input data. For example, information on the temperature at which the literature ERH and viscosity data was taken needs to be reported, as both parameters are temperature dependent. Related, information on the particle size range of the literature data used to build the model should be added, as ERH is known to be a function of particle size.
  - o It remains unclear to me why a training and test data set were created and why not all literature data points were used to derive their eq. 4, as it appears that the presented model is based on a linear regression of the literature data (e.g., L179-181).
  - o A more detailed discussion about the uncertainties of their model is required, as well as of the assumptions made in building the linear regression model. E.g., the difference between the dark pink and light pink shading in Figures 2 and 3 is not sufficiently explained. Moreover, related to my comment above regarding the temperature dependence of both ERH and  $\eta$ , what is the temperature range over which their model can be applied and how does this temperature range compare to temperatures observed in the troposphere. Related, what types of inorganic salts and organics and mixtures thereof is your model applicable to?
  - o There are many formatting flaws within the references in the manuscript that need to be addressed.
    - For example, the spacing between the text and the reference is missing in many instances (e.g., L38 “transmission(Oswin et al. 2022)” vs. “transmission (Oswin et al. 2022)”.
    - Punctuation to mark the end of a sentence sometimes appears before and sometime after a reference, see e.g., L39 vs. L44. This should be made consistent throughout the text.
    - In the bibliography, for several references the same reference appears multiple times, e.g., Lilek and Zuend (2022) “a” and “b” and formatting of references in appears weird in some places, e.g., L298: “(Jonathan P. Reid et al. 2014)”.
- There are several typos and inconsistencies in the manuscript, e.g., L71 “worg”, L127 “aw”, L204 “Tg”, L254 “nucleus formaton”, L256 “link between q to”, to name a few. A thorough round of proof reading and type setting is needed prior to submitting a revised version of the manuscript.
- Section 3.3 “Nucleation kinetics”: This section requires improvement and an overall more detailed discussion.

- I agree that efflorescence is a kinetic process. Are all the ERH data used in their model taken at the same dehumidification rates? This information should be added to the SI tables and discussed in the manuscript.
- L256: “ $-W(T)/KT$  correlates with  $-\log\eta$ ”. Please define and quantify “correlate”. Also, you switch back and forth between  $\log_{10}$  and  $\ln$  logarithm. Please introduce a consistency upon revision.
- Section 4: This section needs to be improved, and statements therein need to be substantiated by a more in-depth discussion. As an example, one case study (L275-278) is used to make claims about SOA particle viscosity. Here, discussing a global perspective, as provided e.g. in the cited reference by Shiraiwa et al. (2017) appears to me more representative. As an other example, to what degree a parameterization as developed here improves representation of aerosols in models predicting air quality and climate (L298-300) remains unclear from the present manuscript.

**Specific:**

- L45: “amorphous organic show no hysteresis”. This statement is wrong, please see e.g., Fig. 7c in the cited Koop et al. (2011) reference.
- L47: “introducing substantial uncertainties...”. Please specify uncertainties, provide examples and appropriate references.
- L58-61: The statement should be supported by references.
- L70: “is widely used”. Since you argue that it is widely used, it would be appropriate to give more than one reference here.
- L120: Please provide more specifics of the AIOMFAC-VISC model and how you used it to determine the viscosities. What are the uncertainties of the “modelled” viscosities used herein?
- L156: Please provide a more detailed discussion for your choice of  $k_{GT}$ .
- Fig. 3: The x-axes are wrong/switched.