

We express our sincere gratitude to the reviewer for the insightful comments and positive assessment of our manuscript, “**Predicting Organic–Inorganic Aerosol Efflorescence Using Thermodynamically Modeled Viscosity**” The reviewers’ suggestions are constructive in enhancing the clarity of our work. We have carefully addressed all comments and will incorporate the corresponding revisions into the updated manuscript. Our detailed, point-by-point responses to the reviewers’ evaluations are provided below.

*1. Page 4, Line 95: The authors note compiling a total of 102 ERH data points, splitting them into 66 for the training set and 36 for the validation set. It would be helpful to briefly clarify the selection criteria used for this split.*

**Response:** We sincerely thank the reviewer for raising this point. We agree that the data division requires further clarification. Our dataset was separated based on the source of data to test the external generalizability of our model. Specifically, the datasets were divided as follows:

Training Set: This set consists of the experimental ERH data from our previous studies, and all corresponding viscosity data were derived using the AIOMFAC model. This dataset was utilized to establish relationship between ERH and other parameters.

Validation Set: This set comprises independent experimental results that either reported both aerosol ERH and viscosity reported or observed no efflorescence. By evaluating the regression model, developed from our own experimental setup, against this entirely independent external dataset, our objective was to demonstrate that the observed relationship is robust and applicable across different laboratory conditions, measurement techniques, and groups.

**Revision: Methods:** *All ERH data in the training were from our previous studies, and all corresponding viscosity data were derived using the AIOMFAC model.*

*2. Page 6, line 156: When calculating the glass transition temperature using the Gordon-Taylor equation, why  $k_{GT}$  is assumed to be 2.5 for all organic-water mixtures?*

**Response:** We thank the reviewer for this insightful comment. We agree that applying a constant Gordon–Taylor parameter ( $k_{GT} = 2.5$ ) across all organic–water mixtures is a generalized assumption. While 2.5 serves as a widely accepted best estimate for

secondary organic aerosol (SOA) components in the absence of experimental data (often estimated within  $2.5 \pm 1.0$ ), the actual value depends on specific solute-water interactions. We have added a brief discussion to the revised manuscript to explicitly acknowledge this simplification and the minor uncertainties it introduces to our model predictions.

**Revision:**

*...It should be noted that assuming a constant Gordon-Taylor parameter ( $k_{GT} = 2.5$ ) for all organic-water mixtures represents a generalized simplification. The  $k_{GT}$  value essentially parameterizes the interaction strength between water and the specific organic solute. Consequently, for organic mixtures with highly complex or varying oxygenated functional groups, the actual  $k_{GT}$  value may deviate from this assigned average (typically estimated within a range of  $2.5 \pm 1.0$ ). This variability introduces a potential, albeit minor, source of uncertainty into the  $T_g$ -based model predictions presented in this study...*

*3. Page 11 line 265: The determination of the viscosity threshold is an excellent finding. It might be beneficial to add one or two sentences contextualizing this value physically—specifically, how this threshold relates to the transition from a liquid to a semi-solid phase in the context of typical atmospheric timescales.*

Response: We sincerely thank the reviewer for this insightful suggestion and for recognizing the value of this finding. We agree that contextualizing the viscosity threshold within atmospheric timescales strengthens the manuscript.

While the characteristic timescale for aerosol microphysical processes and atmospheric residence (e.g., in the marine boundary layer or free troposphere) ranges from days to weeks, which is longer than our laboratory observation window, the fundamental kinetic principles remain the same. Once the particles cross the viscosity threshold into a semi-solid phase, the kinetic barrier to crystallization becomes so immense that efflorescence is highly unlikely to occur, even over extended atmospheric timescales.

Per your suggestion, we have added sentences to the revised manuscript to

physically contextualize this threshold.

**Revision:** *...At this critical viscosity threshold, the aerosol transitions from a liquid to a semi-solid phase, creating an immense kinetic barrier to molecular diffusion. Consequently, the nucleation induction time is prolonged so significantly that efflorescence is effectively inhibited, even over the extended residence times typical of aerosols in the atmosphere (days to weeks)...*

*4. Would other parameters that are not included in the study potentially affect aerosol efflorescence? For example aerosol pH?*

Response: We thank the reviewer for raising this highly relevant question. We agree that aerosol pH is a critical parameter that can potentially influence aerosol phase transitions. In our experimental system, the pH of the droplet solutions ranged from approximately 2.0 to 5.0, which is consistent with the typical pH range observed in global tropospheric aerosols. To investigate the potential impact of pH on aerosol efflorescence, our research group (Sun et al., 2023) analyzed the efflorescence relative humidity (ERH) of various ammonium nitrate (AN)/water-soluble organic compound (WSOC) systems under different pH conditions. The results indicate that the influence of pH is highly dependent on the specific organic species involved. For the AN/sucrose (SUC) and AN/glycerol (GLY) systems, no significant changes in ERH were observed upon acidification to pH 2.0. For the AN/citric acid (CA) system, however, the ERH decreased dramatically to 1–6% when the pH was increased. Under high pH conditions, the formation of partially deprotonated CA and sodium citrate can induce complex interactions, such as ion pairing and hydrogen bonding. These interactions effectively hinder the nucleation and crystallization of AN, thereby influencing its ERH range.

*Page 3, line 78: Change “observational data” to “observational data”.*

*Page 10, line 256: There is an extra “q” between “between” and the following word.*

*Page 6, line 156: Change “assumed” to “assumed”.*

*Page 7, line 180&183: Change “tranning” to “training” and “componets” to “components”.*

*Page 7, line 184&186: Change “altough” to “although” and “nomalized” to “normalized”*

*Page 8, Line 204: In Eq. (6), it should be “ERH”.*

*Figure 3: Some abbreviations in the figure legend (e.g., CA, SUS) are not explained in the caption.*

Response: We sincerely apologize for the spelling and typographical errors that appeared in the initial version of our manuscript and are very grateful to the reviewer for their careful and meticulous examination. We have now corrected all spelling errors throughout the main text, including “observational, assumed, raining, components, although, and normalized.”

### **References**

Sun, J., Hu, Y., Cao, X., Pang, S.-F., Liu, P., Huang, Q., and Zhang, Y.-H.: Role of WSOCs and pH on Ammonium Nitrate Aerosol Efflorescence: Insights into Secondary Aerosol Formation, *Environ. Sci. Technol.*, 57, 20074–20084, <https://doi.org/10.1021/acs.est.3c07603>, 2023.