

We sincerely thank the reviewer for their careful evaluation of our manuscript and for the constructive and insightful comments. We have carefully considered all of the comments and suggestions, and have revised the manuscript accordingly. These revisions have helped us improve the clarity, rigor, and overall quality of the manuscript. Below, we provide a detailed point-by-point response to each comment, and all corresponding changes have been incorporated into the revised manuscript.

Major Comments

1. *Temperature: While particle viscosity is dependent on temperature, the study association between viscosity and ERH is only valid for the temperature at which measurements were made. It is very important to clarify the working temperature throughout the main text, captions and SI.*

Response: We sincerely thank the reviewer for this insightful comment. We have clarified throughout the revised manuscript, including the main text, figure captions, and Supporting Information, that the empirical data and corresponding regression model are based on measurements conducted at 298 K. We have also explicitly stated the temperature limitation of the model and emphasized that its applicability is restricted to standard room-temperature conditions.

Revision: *It should be emphasized that the empirical data utilized to derive this regression model were obtained at a constant temperature of 298 K. Given the well-established temperature dependence of both ERH and viscosity, the applicability of the current model is limited to standard room temperature conditions (298±10K). Extrapolating this model to broader temperature regimes would necessitate further temperature-specific calibrations.*

2. *Section 2.1: How were data points chosen to make the training set? Were they randomly selected?*

Response: We sincerely thank the reviewer for raising this important question. The data points were not randomly selected to construct the training set. Instead, the dataset split was performed according to data source, with the specific aim of evaluating the external generalizability of the model rather than its performance under a random internal split.

The training set consisted of the experimental ERH data obtained in our own laboratory, for which the corresponding viscosity values were derived using the AIOMFAC model. This dataset was used to establish the relationship between ERH and viscosity-related parameters. The validation set was composed of independent datasets collected from the literature, including studies that reported both aerosol ERH and viscosity, as well as cases where no efflorescence was observed.

We chose this source-based separation intentionally because it provides a more rigorous test of model robustness. By validating the regression developed from our own dataset against an entirely independent external dataset, we aimed to examine whether the identified relationship remains applicable across different research groups, experimental conditions, and measurement approaches.

3. Section 3.2: Have you tried another form of regression model? What is the rationale for picking the linear regression model as the choice?

Response: We sincerely thank the reviewer for this insightful comment. In the present study, the linear regression model was selected primarily because our objective was to establish a simple, interpretable, and empirically robust relationship between the variables within the available dataset. Compared with more complex nonlinear forms, a linear model offers clearer physical interpretability, reduces the risk of overfitting given the limited data size, and facilitates direct application and comparison across independent datasets.

In addition, the observed trend in our dataset did not warrant the introduction of a more complex functional form at this stage. Therefore, we chose the linear regression model as a first-order empirical description of the relationship. We agree that exploring alternative regression forms may be valuable in future work, particularly when a larger and more diverse dataset becomes available.

4. Conclusions and Implications: When discussing the implications for the ambient

aerosols, could you please discuss in the border context including places other than China?

Response: We sincerely thank the reviewer for the valuable comment. We agree that the previous discussion was too limited in scope and did not sufficiently substantiate the broader atmospheric implications of our results. In the revised manuscript, we have expanded this section by incorporating a more representative global perspective based on Shiraiwa et al. (Shiraiwa et al., 2017), who showed that SOA phase state varies strongly with relative humidity and temperature on the global scale, with liquid SOA prevailing in humid tropical and polar regions, semi-solid SOA in mid-latitudes, and solid SOA over dry lands in the planetary boundary layer, while SOA in the middle and upper troposphere are predicted to be mostly glassy.

Revision: *From a broader atmospheric perspective, the implications of aerosol phase state should be considered on regional to global scales. Shiraiwa et al. (Shiraiwa et al., 2017) showed that SOA phase state varies strongly with relative humidity and temperature, with SOA in the planetary boundary layer being predominantly liquid in humid tropical and polar regions, semi-solid in mid-latitudes, and solid over dry lands, while SOA in the middle and upper troposphere are expected to be mostly glassy.*

Minor Comments

1. Lines 64 – 67: The current description is too vague. Could you elaborate more how different factors listed here influence the efflorescence?

Response: We sincerely thank the reviewer for this helpful comment. Component diffusion controls the mobility of ions and molecules and therefore affects the rate at which crystalline nuclei can form and grow; higher viscosity generally suppresses diffusion and delays efflorescence. Chemical composition, including solute concentration, solubility, and vapor pressure, influences the degree of supersaturation and thus the thermodynamic driving force for crystallization. In addition, molecular interactions within the particle phase can either facilitate or hinder structural rearrangement required for nucleation, while interfacial properties such as surface tension affect the nucleation energy barrier by altering the energetic cost of forming a new phase interface.

Revision: *Efflorescence is a kinetically-controlled process that requires overcoming a nucleation energy barrier. This process is influenced by several factors. Component diffusion controls the mobility of ions and molecules and therefore affects the rate at which crystalline nuclei can form and grow; higher viscosity generally suppresses diffusion and delays efflorescence. (Ji et al., 2017; Mikhailov et al., 2009) Chemical composition, including solute concentration, solubility, and vapor pressure, influences the degree of supersaturation and thus the thermodynamic driving force for crystallization. (Cohen et al., 1987; Gupta et al., 2015) In addition, molecular interactions within the particle phase can either facilitate or hinder the structural rearrangement required for nucleation, while interfacial properties such as surface tension affect the nucleation energy barrier by altering the energetic cost of forming a new phase interface. (Davis et al., 2015; Ma et al., 2021; Mikhailov et al., 2009)*

2. Lines 74–76: *The sentence “Under such conditions, aerosols may shift from ... glass transition temperature (T_g)” is unclear. Could you please rephrase it? I do not see how the efflorescence behavior shift to amorphous phase transitions.*

Response: We sincerely thank the reviewer for this helpful comment. We agree that the original sentence was unclear and could incorrectly imply that efflorescence behavior directly “shifts” to an amorphous phase transition. Our intention was to convey that, under conditions of high viscosity and slow molecular diffusion, crystallization may become kinetically hindered or even suppressed. In such cases, aerosol particles can remain in an amorphous semisolid or glassy state rather than undergoing efflorescence, and their phase-state evolution is then more appropriately discussed in terms of the glass transition temperature (T_g). We have therefore rephrased this sentence in the revised manuscript to make this distinction clearer.

Revision: *Under such conditions, efflorescence may be kinetically suppressed because of limited molecular diffusion, causing aerosol particles to remain in an amorphous semisolid or glassy state; in this case, their phase-state evolution is more appropriately characterized with reference to the glass transition temperature (T_g).*

3. *Could you please specify the temperature for the data points collected? Were they*

measured in the same range? I assume temperature can affect efflorescence. Also, temperature is one of the inputs in AIOMFAC.

Response: We sincerely thank the reviewer for this insightful comment. We have clarified throughout the revised manuscript, including the main text, figure captions, and Supporting Information, that the empirical data and corresponding regression model are based on measurements conducted at 298 K. We have also explicitly stated the temperature limitation of the model and emphasized that its applicability is restricted to standard room-temperature conditions.

Revision: *It should be emphasized that the empirical data utilized to derive this regression model were obtained at a constant temperature of 298 K. Given the well-established temperature dependence of both ERH and viscosity, the applicability of the current model is strictly limited to standard room temperature conditions (298±10K). Extrapolating this model to broader temperature regimes would necessitate further temperature-specific calibrations.*

4. Eq 2: The calculation of C^0 is based on the parameterization of Li et al., 2016. Please note that this is not one parameterization for C^0 . To ensure the robustness of the claims in the manuscript, I would suggest including other C^0 parameterizations to evaluate how the result of T_g and so the association between ERH and T_g will change.

Response: We sincerely thank the reviewer for this valuable comment. We agree that the parameterization of C^0 is not unique, and that different parameterizations may lead to some variation in the calculated T_g values. In the present study, we adopted the parameterization of Li et al. (Li et al., 2016) because it is based on extensive evidence showing a close relationship between volatility and viscosity, and it develops parameterizations for predicting T_g as a function of C^0 using data from more than 2000 compounds. In addition, functional-group contribution approaches are widely used to predict C^0 (Capouet and Müller, 2006; Pankow and Asher, 2008), meaning that the use of C^0 as a predictor of T_g can also indirectly account for molecular-structure effects. Therefore, the parameterization of Li et al. provides a well-established and internally consistent basis for our C^0 and T_g calculations.

At the same time, because our ultimate purpose is to evaluate the relationship between ERH and T_g , we assessed the robustness of this relationship not by introducing additional C^0 parameterizations, but by applying alternative and independent methods for estimating T_g . Specifically, in the revised manuscript we included the framework of Armeli et al. (Armeli et al., 2023), which provides several prediction modes based on different types of input information, including Functional Group Mode (FGM), SMILES Mode (SM), and their corresponding variants with melting temperature (T_m) as an additional input.

The results obtained from these alternative T_g estimation approaches are summarized in Tables S4–S7. Although the absolute T_g values differ somewhat among methods, the overall association between ERH and T_g remains consistent.

Revision: *Multiple parameterizations exist for estimating the glass transition temperature (T_g) of organic compounds, utilizing predictors such as chemical structure, volatility, or melting point (T_m) (Armeli et al., 2023; Galeazzo and Shiraiwa, 2022; Koop et al., 2011; Li et al., 2020). Recent machine-learning models based on chemical structure do not strictly require T_m information, though including it as an optional parameter can further improve predictive accuracy (Armeli et al., 2023). The framework proposed by Armeli et al. comprises four distinct modes, contingent upon the specific input parameters available for a given compound: the Functional Group Mode (FGM) and the SMILES Mode (SM). Each mode features a variant that includes melting temperature as an additional feature (FGM with T_m ; SM with T_m) and a version that operates without it (FGM no T_m ; SM no T_m). The T_g values and parameters calculated according to the methodology of Armeli et al. are summarized in Tables S4 – S7.*

The results, presented in Figure 1c, demonstrate that neither methodology yields a statistically significant correlation. Furthermore, analyses employing alternative FGM and SMILES models for T_g estimation (Figure S2), and the T_g for organic-inorganic were also investigated (Figure S2). Similarly, no discernible correlation with ERH was observed.

5. Line 186: Please include the description of the normalized aerosol ERH in the main

text.

Response: We sincerely thank the reviewer for this helpful suggestion. In the revised manuscript, we have added a definition of the normalized aerosol ERH, expressed as ERH/ERH_{Ing} , where ERH_{Ing} represents the efflorescence relative humidity of the corresponding inorganic salt system. We have also clarified that this normalized parameter was introduced to evaluate the influence of organic components on aerosol efflorescence behavior relative to the inorganic reference. Accordingly, the relevant description has now been incorporated into the main text

Revision: *We found that although aerosol efflorescence occurs due to the crystallization of aqueous inorganic salt, the correlation between aerosol viscosity and the normalized aerosol ERH defined as ERH/ERH_{Ing} (where ERH_{Ing} is the efflorescence relative humidity of the corresponding inorganic salt system) was even weaker ($R^2 = 0.59$, Figure S3).*

6. Line 200: *What does a Sig.F change mean?*

Response: We sincerely thank the reviewer for pointing out this lack of clarity. “Sig.F change” refers to the significance of the F-change test, which evaluates whether the inclusion of additional predictors leads to a statistically significant improvement in model performance compared with the simpler baseline model.

7. Fig 3: *It looks like the x-axis labels have been swapped between the two subplots.*

Response: Thank you for your careful review. We have revised the figure accordingly: the x-axis labels for panels (a) and (b) have been corrected, and the y-axis for panel (a) now reads “Predicted ERH”.

8. Lines 247-248: *Could you elaborate on “viscosity plays a central role in governing nucleation rates”? The current sentence is unclear.*

Response: We sincerely thank the reviewer for this helpful comment. We agree that the original statement was insufficiently supported by references and could be made more precise. In the revised manuscript, we have removed the overly general sentence

“viscosity plays a central role in governing nucleation rates” and rewritten this part to more clearly describe the physical meaning of the kinetic term in the nucleation expression.

Revision: *In highly viscous aerosols, mass transfer limitations significantly alter nucleation dynamics, making nucleus formation the rate-limiting step (Ji et al., 2017; Wang et al., 2017). Turnbull and others showed that the nucleation rate can be expressed as the product of a pre-exponential factor and two thermally activated rate terms (Hollomon and Turnbull, 1953; Kelton et al., 1983; Turnbull, 1969):*

$$\tau_x^{-1} = v \exp \left[\frac{-W(T)}{kT} \right] \exp \left[\frac{-\Delta G(T)}{kT} \right] \quad (9)$$

where $W(T)$ represents the kinetic barrier associated with atomic rearrangement, and $\Delta G(T)$ is the thermodynamic free energy barrier for nucleus formation. The term $\exp(W(T)/kT)$ represents the thermally activated atomic rearrangement rate (i.e., atomic mobility) in liquids and is inversely related to viscosity (η^{-1}). Here, we introduce an approximate proportional relationship between the kinetic barrier term and the logarithm of viscosity:

$$\frac{-W(T)}{kT} = -A \log_{10} \eta \quad (10)$$

where A is a constant linking $-W(T)/kT$ with $-\log_{10} \eta$. Substituting this relation into Eq. (9) and taking \log_{10} gives:

$$\log_{10}(\tau_x^{-1}) = -A \log_{10} \eta + \log_{10} v - \frac{\Delta G(T)}{2.303kT} \quad (11)$$

The empirical relationship between ERH and aerosol viscosity (Eq.6) is consistent with Eq. 11, indicating that $\log_{10} \eta$ serves as a proxy for the kinetic energy barrier $W(T)$, while ERH reflects the effective nucleation rate. The intercept achieved in Eq.6 can therefore be associated with $\log_{10} v - \frac{\Delta G(T)}{2.303kT}$.

9. Lines 254-256: Could you provide references to the sentence “The term exp... to viscosity.”

Response: Please refer to the reply to Comment 7.

Technical Comments

Please ensure the consistency of formatting throughout the manuscript, for example:

- 1. lines 38 -39: space should be added before “(Oswin et al., 2022)” and “(Reid et al., 2018)” line 42. (Novo et al., 2021) should be placed before the full dot.*
- 2. Lines 62-63: Please add references for “While laboratory studies have... remains limited”*
- 3. Line 71: It is supposed to be “ ω_{org} ” instead of “ ω_{org} ”.*
- 4. Line 72: It is better to use “highly viscous” rather than “high-viscosity”.*

Response 1-4: We thank the reviewer for careful and detailed comments. The above question has been revised.

- 5. Line 76: What direct and reliable measurement? Composition? Hygroscopicity?*

Response: We sincerely thank the reviewer for pointing out this ambiguity. Here, “direct and reliable measurement” specifically refers to the direct measurement of aerosol viscosity under atmospherically relevant conditions, rather than to composition or hygroscopicity. Our intention was to emphasize that viscosity measurements of atmospheric aerosol particles remain challenging, particularly because particle water uptake can continuously alter aerosol composition, phase state, and molecular mobility. We have revised the sentence to make this point explicit.

Revision: *However, direct and reliable measurements of the viscosity of atmospheric aerosols under atmospherically relevant conditions remain challenging (Fitzgerald et al., 2016; Hosny et al., 2016), particularly because water uptake can continuously modify particle composition, phase state, and molecular mobility. (Sheldon et al., 2023; Zobrist et al., 2008) This has resulted in limited observational data and motivated the use of thermodynamic models, such as AIOMFAC, to predict the viscosity of organic–inorganic mixed aerosols (Lilek and Zuend, 2022)*

- 6. Line 103: It should be clear that x_{org} is the organic molar fraction in the solute. Also, in line 124, should it be x_{org} ?*

Response: We sincerely thank the reviewer for pointing out this lack of clarity. In our calculations, x_{org} refers to the mole fraction of the organic component in the solute

mixture, where the solute includes both organic compounds and inorganic salts. We used this definition because the AIOMFAC input scheme is based on mole fractions for the solute components.

Revision: *Accordingly, viscosity calculations were performed by specifying the functional groups of the organic components, inorganic salt composition, organic-to-inorganic ratio (OIR, molar ratio of organic compounds to inorganic salts, excluding water), and a series of organic molar fractions in the solute mixture, x_{org} , where the solute consists of organic compounds and inorganic salts, ranging from 0.01 to 0.99 in increments of 0.01.*

7. Tables S1 and S2: The abbreviation of OIR is missing. Was the OIR calculated in mass or in moles? Could you provide the organic molar fraction in the tables?

Response: We sincerely thank the reviewer for pointing out these issues. In the revised manuscript, we have explicitly defined OIR as the organic-to-inorganic ratio. The OIR used in this study was calculated on a molar basis, i.e., as the molar ratio of organic compounds to inorganic salts, excluding water.

To improve clarity, we have added the corresponding organic mole fraction (x_{org}) to the relevant tables where this parameter is involved. However, we did not add x_{org} to Table S2, because the data in Table S2 belong to the validation dataset and do not involve the calculation or use of x_{org} .

8. Line 127: aw was only used two times for water activity. This abbreviation of water activity is redundant

Response: We sincerely thank the reviewer for this comment. We have removed this abbreviation in the revised version.

Reference

Armeli, G., Peters, J.-H., and Koop, T.: Machine-Learning-Based Prediction of the Glass Transition Temperature of Organic Compounds Using Experimental Data, ACS Omega, 8, 12298–12309, <https://doi.org/10.1021/acsomega.2c08146>, 2023.

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