We sincerely thank the reviewer for their careful review of our manuscript and for the overall positive feedback. We have addressed all comments from the reviewer with point-by-point responses with corresponding revisions made to the manuscript. Our detailed response and revisions are provided below.

General comments:

1. Introduction (lines 43, 64): Please define or explain the meaning of efflorescence and deliquescence more carefully. Note that often not the whole particle undergoes deliquescence; rather, it is usually a solid (crystalline) salt or hydrate that does, while the remaining particle may be in an amorphous liquid or semi-solid state. Related to the above and the statement on line 64: "Efflorescence is a kinetically-controlled process that requires overcoming a nucleation energy barrier, …", it should be further clarified under what conditions efflorescence can occur, as well as efflorescence due to what material crystallizing. In mixed organic-inorganic aerosols, it is typically not the case that the whole particle crystallizes, only a specific portion of inorganic and sometimes organic materials may crystallize. Furthermore, efflorescence is not entirely kinetically controlled. If the (aqueous) solution is not supersaturated thermodynamically with respect to the solid crystalline phase that may form, efflorescence is thermodynamically prohibited. The fact that some degree of supersaturation is needed to crystallize inorganic materials like ammonium sulfate or sodium chloride, is well known. For example, see the works by Marcolli and Krieger (2006, https://pubs.acs.org/doi/10.1021/jp0556759), Hodas et al. (2016, www.atmos-chem-phys.net/16/12767/2016/), Bouzidi et al. (2020, https://doi.org/10.1016/j.scitotenv.2022.153010).

Response: We sincerely thank the reviewer for improving the accuracy for our discussions of efflorescence and deliquescence, which has significantly deepened our understanding of these phenomena. We have added clarifications following your suggestion and cited the references to support our discussion.

Revision:

"It is important to note that, in internally mixed organic–inorganic aerosols, the onset of phase transitions such as deliquescence and efflorescence typically occur only in

specific components rather than the entire particle. For example, crystalline inorganic salts or hydrates may undergo deliquescence or crystallization, while the remaining particle matrix can persist in an amorphous, semi-solid, or aqueous state (Hodas et al., 2016)"

"Furthermore, thermodynamic constraints exists for efflorescence. Crystallization can only occur when the aqueous phase becomes supersaturated with respect to the relevant solid phase; otherwise, efflorescence is thermodynamically inhibited. Once supersaturation is achieved, nucleation and crystal growth proceed through kinetically controlled processes (Bouzidi et al., 2022; Marcolli and Krieger, 2006)."

2. The study by Hodas et al. (2016) describes a thermodynamics-based method to model the conditions of salt crystallization and the associated ERH. Did the authors consider such thermodynamics-based methods? There is likely a close correlation between relative humidity (RH) and viscosity, as well as RH and degree of salt supersaturation relative to the ion activity product at saturation. Such effects and alternatives to predicting ERH should at least be discussed.

Response: We sincerely thank the reviewer for this insightful suggestion and for highlighting the work by Hodas et al. (2016). We agree that thermodynamics-based frameworks, particularly those based on the ion activity product (IAP), provide a physically grounded basis for predicting salt crystallization and ERH.

In our framework, we assume that aerosols near ERH exist at relatively high ionic strength, consistent with the concentrated conditions expected at low water activity. Under these conditions, we estimate the dynamic viscosity of organic–inorganic mixtures and focus on its role in modulating efflorescence. At the same time, we acknowledge that thermodynamics provides a complementary control: the degree of supersaturation (e.g., relative to the IAP) determines whether crystallization is thermodynamically feasible. In the absence of supersaturation, efflorescence cannot occur. Once this condition is met, the observed ERH is governed by kinetic limitations associated with nucleation and crystal growth, for which aerosol viscosity serves as an effective proxy.

We have now added a discussion of thermodynamics-based approaches and their relationship to our viscosity-based parameterization in the revised manuscript.

Revision: *"Thermodynamics-based approaches, such as that described by Hodas et al. (2016), predict salt crystallization and ERH based on the degree of supersaturation relative to the ion activity product (IAP). These frameworks capture the fundamental requirement that crystallization can only occur when the aqueous phase becomes thermodynamically supersaturated with respect to the relevant solid phase.*

In this study, we consider aerosol conditions near ERH, where particles are expected to exhibit high ionic strength due to reduced water content. Under these conditions, we evaluate aerosol viscosity and its influence on phase transitions. Efflorescence is therefore interpreted as a coupled thermodynamic–kinetic process: while supersaturation provides the necessary driving force for crystallization, the actual onset of efflorescence is modulated by kinetic barriers associated with nucleation and crystal growth. In this context, aerosol viscosity plays a key role by limiting molecular diffusion and ion mobility.

Thus, thermodynamic and kinetic perspectives are complementary: thermodynamic conditions determine whether crystallization is possible, whereas viscosity provides a practical proxy for the kinetic limitations that govern when crystallization occurs."

3. Citations/References: several papers seem to be cited more than once and given different reference entries although they are referring to the exact same paper. For example, there is only one Li et al. (2020) paper but these and others are referenced with a,b postfixes. This needs to be fixed throughout the manuscript.

Response: We sincerely thank the reviewer for their careful reading. We have thoroughly checked the in-text citations and the reference list throughout the manuscript. All duplicate reference entries referring to the exact same paper, including the specific example of Li et al. (2020), have been carefully merged and corrected. The unnecessary "a" and "b" postfixes have been systematically removed to ensure the accuracy of the bibliography and strict compliance with the journal's formatting guidelines.

4. If high viscosity suppresses efflorescence or strongly affects ERH, this may mean that measured ERH values can be affected by the protocol followed during experiments, such as the rate of dehumidification and time scale of the experiment overall. Efflorescence may still occur at nonzero relative humidity, but the experimenters may not have waited long enough. Please add some discussion of this aspect, e.g. how experimental time scales compare to average aerosol lifetime in the troposphere.

Response: We sincerely thank the reviewer for this insightful comment regarding the potential influence of experimental timescales on observed ERH. We agree that, for highly viscous particles, delayed nucleation could lead to underestimation of efflorescence if the observation window is insufficient.

To address this, we have systematically compared our laboratory timescales with atmospheric timescales and evaluated the delay in efflorescence based on nucleation induction times.

In the experiments included in our dataset, a full dehydration cycle from >80% RH to <10% RH typically lasted 60–90 minutes, with a gradual RH change (~0.067% s^{-1}) to ensure equilibrium conditions, resulting in a whole experiment period of several hours.

Regarding atmospheric timescales, Anon et al. (2000) pointed out that the characteristic timescale for aerosol microphysical processes ranges from days to weeks, which is generally longer than the residence time of aerosols in typical atmospheric compartments (such as the marine boundary layer and the free troposphere). We acknowledge that our laboratory observation time (several hours) is substantially shorter than these atmospheric residence times. However, as demonstrated by Wang et al. (2017) for sufficiently viscous systems (e.g., OIR > 1:1), the kinetic barrier to crystallization is so immense that efflorescence is highly unlikely to occur even on extended atmospheric timescales.

Therefore, we interpret the suppression of efflorescence in high-viscosity particles as a physically meaningful kinetic limitation rather than an artifact of experimental duration. We have added a discussion to clarify this point in the revised manuscript.

Revision: "*For dataset included in this study, experiments were conducted over several hours with slow RH changing rate (< 0.1%/s) to ensure near-equilibrium conditions. Although these timescales are shorter than typical atmospheric aerosol lifetimes (days to weeks), previous studies have shown that nucleation induction times in highly viscous systems can be extremely long due to diffusion limitations (Wang et al., 2017). Therefore, the suppression of efflorescence at high viscosity reported in the reference is likely a fundamental kinetic constraint rather than an artifact of limited experimental duration.*"

5. For all figures and related tables: the temperature (ranges) of the data shown should be indicated. Both ERH and viscosity depend on temperature. It is therefore also not clear whether the derived regression model applies only to room temperature (assuming most data are for about 293–298 K) or more broadly.

Response: We thank the reviewer for pointing out this critical detail. Because both ERH and viscosity are temperature-dependent, explicitly specifying the temperature condition is crucial for the rigorous interpretation and practical application of the data presented in this study. Based on your constructive suggestion, we have updated all relevant figures and tables in the manuscript. The specific temperature condition (298 K) is now explicitly indicated in the captions or footnotes of all applicable figures (Figure 2) and tables (Tables S1 and S2). Furthermore, we have clarified in the revised text that the derived regression model was established strictly based on data collected at a constant temperature of 298 ± 10K. Therefore, the current model is specifically applicable to standard room temperature conditions (298 ± 10K). We have added a relevant statement to the manuscript to ensure that readers clearly understand this boundary of applicability.

Revision: *"Moreover, the empirical data utilized to derive the regression model in this study were all obtained within a temperature range of 298 ± 10K."*

Specific comments:

1. Line 46–48: To add to the statements there, for clarity and completeness, aerosol particles consisting of more than a single phase may have crystalline as well as amorphous phases.

Response: We have added the following statement to Lines 46–48: “Moreover, the empirical data utilized to derive the regression model in this study were all obtained within a temperature range of 298±10 K.”

2. Lines 94–96: There seem to be additional relevant datasets that the authors have not considered, e.g. Marcolli and Krieger (2006, https://pubs.acs.org/doi/10.1021/jp0556759) and Ciobanu et al. (2010, https://pubs.acs.org/doi/10.1021/jp103541w). Ciobanu et al. (2010), in their Fig. 1 and Table 1, suggest that the ranges of the ERH of AS in particles may depend on particle diameter (for a range of similar temperatures). Their paper further discusses possible influence of phase separation on the ERH of AS, which would mean AS efflorescence may be affected by the presence of an organic coating in core-shell aerosols. In such cases, an inner aqueous AS-rich phase may show lower ERH independent of viscosity effects since the viscosity of the inner aqueous AS-rich phase would be similar to that of organic-free aqueous AS particles, which were shown to effloresce at a higher relative humidity in that study. Such findings are therefore questioning the reliability of predicting ERH solely based on viscosity and should be discussed in the context of the current studies findings.

Response: We sincerely thank the reviewer for the highly relevant datasets and for highlighting the critical physical mechanisms underlying efflorescence. We acknowledge that our initial literature review was not comprehensive enough in this regard, and we greatly appreciate your constructive input. Predicting ERH based solely on bulk viscosity has limitations.

In mixed organic-inorganic systems, liquid-liquid phase separation (LLPS) frequently results in core-shell morphologies. In such cases, the inner aqueous AS-rich core may maintain a low, liquid-like viscosity, yet the ERH can still be significantly suppressed due to the organic coating altering the interfacial tension or acting as a physical barrier. This suppression occurs independently of the inner phase's viscosity.

We do not intend to claim that bulk viscosity is the sole determining factor for ERH. Rather, it serves as a macroscopic proxy that is particularly dominant under certain mixing states. To address your concerns and improve the scientific rigor of our paper, we have added a dedicated discussion in the revised manuscript.

Revision: *"while our results demonstrate a clear empirical correlation between viscosity and ERH, it is crucial to acknowledge that predicting ERH solely based on bulk viscosity has inherent limitations. As highlighted by previous studies (Ciobanu et al., 2010; Marcolli and Krieger, 2006), in complex organic-inorganic systems, liquid-liquid phase separation (LLPS) frequently occurs, forming core-shell morphologies. In such core-shell aerosols, the inner aqueous inorganic-rich phase may maintain a low, liquid-like viscosity comparable to that of pure inorganic droplets, yet exhibit a suppressed ERH. This suppression can arise because the organic coating alters the interfacial energy or acts as a physical barrier, independent of the inner core's viscosity (Ciobanu et al., 2010). Therefore, while bulk viscosity acts as a critical kinetic constraint, the observed ERH is ultimately governed by a complex interplay of various factors including viscosity, particle size, phase morphology etc."*

3. Line 102: related to the general comments: "aerosol ERH" should likely be rephrased to refer to the ERH of a specific electrolyte/salt that can solidify in the aerosol. The remaining organic-rich aerosol phase may contain water and a small amount of dissolved ions from the salt components, hence it is misleading to talk of an overall "aerosol ERH". In a multicomponent organic-inorganic particle, several distinct ERH may exist due to different salts crystallizing at different RH thresholds.

Response: We sincerely thank the reviewer for this comment regarding the microscopic physical nature of efflorescence. We agree that, in internally mixed organic–inorganic particles, different inorganic salts may crystallize at distinct relative humidities, and the remaining organic-rich phase can retain water and dissolved ions. Therefore, referring to a single "aerosol ERH" may oversimplify the underlying complexity.

In this study, we retain the term “aerosol ERH” as a macroscopic, operational parameter. The crystallization of dominant inorganic components typically induces substantial changes in particle morphology, water content, optical properties, and apparent phase state. As such, we use “aerosol ERH” to represent the overall transition of the particle system from a predominantly liquid to a mixed-phase or semi-solid/solid state.

We also note that efflorescence is not a discrete point but occurs over a relative humidity range. To enable quantitative model development, the ERH used here is defined as the mean value of the observed transition range. We have clarified this definition in the revised manuscript.

Revision: *“In multicomponent organic–inorganic particles, multiple efflorescence relative humidities may exist due to different salts crystallizing at distinct RH levels, and efflorescence typically occurs over a RH range. In this study, “aerosol ERH” is defined as the mean value of this transition range, serving as a simplified but effective macroscopic parameter for the overall phase transition of the particle.”*

4. Line 103: water (an inorganic) is not included here as part of n_inorg in the definition of x_org, right?

Response: Yes, water is not included in the calculation of n_{inorg}. The term n_{inorg} strictly refers to the number of moles of the inorganic salt(s) in the system. Therefore, x_{org} represents the molar fraction of the organic component relative to the total dry solutes (organic species plus inorganic salts), which directly corresponds to the molar ratio of organic to inorganic components prepared in our experiments.

5. Lines 110–115: This description of AIOMFAC-VISC seems to be incomplete. Looking both at the description of “Mixture viscosity prediction with AIOMFAC-VISC” on the https://aiomfac.lab.mcgill.ca/help.html page, as well as Lilek and Zuend (2022), the absolute rate theory approach is seemingly only used for the viscosity of aqueous inorganic phases, but not for aqueous organics; see Song et al. (2021, https://doi.org/10.5194/acp-21-10215-2021) and Lilek and Zuend (2022). Also, the

Gervasi et al. reference on line 114 is not properly cited in the manuscript.

Response: We sincerely thank the reviewer for their careful reading and for pointing out this inaccuracy. We agree that our original description was imprecise and have added additional clarification about the use of AIOMFAC-VISC.

As the reviewer noted, the absolute rate theory (Eyring's approach) within AIOMFAC-VISC is exclusively applied to the aqueous inorganic (electrolyte) phase. The aqueous organic phase, relies on a combinatorial-activity-weighted approach developed by Gervasi et al. (Gervasi et al., 2020). By default, the viscosity of mixed organic–inorganic phases is estimated by the "aquelec" approach (in AIOMFAC-web), which represents the effects of ions on viscosity by computing first the viscosity of the aqueous electrolyte mixture (excluding organics) and then uses that sub-system's viscosity to represent a modified viscosity of water in the calculation of viscosity from the (electrolyte-free) sub-system via the aqueous organic viscosity model.

Validation against experimental data (e.g., Figure 11 in Lilek and Zuend, 2022) (Lilek and Zuend, 2022)has demonstrated that the "aquelec" approach accurately characterizes the viscosity of internally mixed aerosols such as sucrose-nitrate systems, establishing it as a robust tool for our predictions.

Revision: *"Aerosol viscosity at the corresponding ERH and 298 K was estimated using the Aerosol Inorganic-Organic Mixtures Functional Groups Activity Coefficients (AIOMFAC) model.(Zuend et al., 2008, 2011). AIOMFAC is a thermodynamics-based group-contribution framework developed for aqueous organic-inorganic mixtures. To predict the viscosity of complex organic-inorganic mixtures, the AIOMFAC-VISC framework conceptually divides the mixture into two distinct sub-systems. For the aqueous inorganic (electrolyte) sub-system, it employs a semi-empirical model based on Eyring's absolute rate theory, which relates viscosity to the molar Gibbs energy of activation for viscous flow (Δg^*) taking into account temperature, ion activities, and ionic strength. Conversely, for the aqueous organic sub-system, the model relies on a combinatorial-activity-weighted approach based on functional group contributions (Gervasi et al., 2020). Adopting the UNIFAC concept, AIOMFAC segments organic molecules into functional subgroups, treating the liquid mixture as a 'solution of* *groups' to manage chemical complexity(Fredenslund et al., 1975; Hansen et al., 1991) In our study, the viscosity of mixed organic-inorganic phases was estimated using the default "aquelec" (electrolyte-aware water) approach in AIOMFAC-web. This method represents the effects of ions by first computing the viscosity of the aqueous electrolyte mixture (excluding organics), and then using that sub-system's viscosity to represent a modified viscosity of water in the calculation of the electrolyte-free organic sub-system. Validation against experimental data has demonstrated that the "aquelec" approach accurately characterizes the viscosity of internally mixed aerosols such as sucrose-nitrate systems, establishing it as a robust tool for our predictions.(Lilek and Zuend, 2022; Zuend et al., 2011). Validation against experimental viscosity data for sucrose–nitrate systems demonstrated that the "aquelec" approach accurately characterizes the viscosity of internally mixed organic–inorganic aerosols, establishing AIOMFAC-VISC as a robust tool for predicting atmospheric aerosol viscosity when experimental measurements are unavailable.(Lilek and Zuend, 2022)"*

6. Line 117: the Fredenslund et al., 1975; Hansen et al., 1991; references are about the UNIFAC concept mentioned in the previous sentences, not about mixing approaches.

Response: We appreciate the reviewer for catching this citation misplacement. We have corrected this error in the revised manuscript. The references (Fredenslund et al., 1975; Hansen et al., 1991) have been moved to the preceding sentence so that they correctly correspond to the UNIFAC concept.

7. Line 121: since this is about sucrose-nitrate systems, the reference to be cited should likely be Song et al. (2021, https://doi.org/10.5194/acp-21-10215-2021) not Lilek and Zuend (2022)?

Response: We sincerely thank the reviewer for highlighting the highly relevant experimental work by Song et al. (2021) regarding sucrose-nitrate systems. Our original citation of Lilek and Zuend (2022) was intentional and serves a specific methodological purpose in our manuscript. Since we utilized the default "aquelec" approach in AIOMFAC-web, citing Lilek and Zuend (2022) is essential for validating our methodological choice and its reliability. We have now also cited Song et al. (2021) in our revised manuscript.

8. Line 123: Is OIR including or excluding water? Also, clarify whether it is mass- or mole-based.

Response: We thank the reviewer for this helpful clarification request. The OIR (organic-to-inorganic ratio) in this study is defined as the molar ratio of organic compounds to inorganic salts, excluding water. We have clarified this definition in the revised manuscript.

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),…"*

9. Line 127: What is 1 CH OH? Clarify this notation. Presumably also notation like CH2 is really CH_2 with subscripts?

Response: We thank the reviewer for pointing this out. In the context of the AIOMFAC model's group-contribution framework, "1 CH OH" refers to one specific functional subgroup consisting of a methine group (CH) bonded to a hydroxyl group (OH). We have corrected the formatting of chemical symbols such as "CH_2" to ensure proper subscript notation, thereby improving the clarity of the presentation.

10. Line 136: "volatility melting point" wording issue

Response: The correction has been incorporated into the text accordingly.

11. Line 156: A note perhaps worthy of mentioning in the manuscript: in addition to the outlined process for T_g(omega_org), one could also determine T_g of the mixture as a whole, including the effects from inorganic ions on T_g, by applying the viscosity predicted by AIOMFAC-VISC together with an assumption of the fragility parameter to the Vogel–Tammann–Fulcher equation (e.g. see Eqs. 6,7 of DeRieux et al. (2018,

https://doi.org/10.5194/acp-18-6331-2018). However, the pure-component viscosity model for organics employed in AIOMFAC-web is probably less skillful than other methods, such as that by Armeli et al. (2023). However, for the purpose of the regression model, the usefulness of the T_g of the mixture predicted from mixture viscosity overall may be negligible since it would likely be redundant with just predicting mixture viscosity as already done.

Response: We sincerely thank the reviewer for this highly constructive comment. We fully agree with your perspective. Following your excellent suggestion, we have added a note to the revised manuscript discussing this alternative approach.

Revision: *"In addition to the approach used here for estimating Tg(*ω_{org}*), the glass transition temperature of the entire mixture, including contributions from inorganic ions, could also be derived from the predicted mixture viscosity. This can be achieved by combining AIOMFAC-VISC viscosity predictions with an assumed fragility parameter within the Vogel–Tammann–Fulcher framework (DeRieux et al., 2018). However, the pure-component viscosity parameterization for organics implemented in AIOMFAC-web may be less accurate than more recent approaches (Armeli et al., 2023). Moreover, for the purposes of the present regression model, estimating Tg from mixture viscosity is likely redundant, as it does not provide additional predictive power beyond viscosity itself."*

12. Figure 1: It would be of interest to show supplementary figure(s) where these scatter plots are shown separately for some of the different inorganic salts, e.g. for AS only, since the ERH of the binary aqueous salt solutions already differ and those may not be a function of viscosity. In that context, is there are clearer correlation obtained for specific salts or not?

Response: We sincerely thank the reviewer for this insightful suggestion. Per your recommendation, we have generated supplementary scatter plots separating the data by ammonium sulfate mixtures, as shown in the figure below. Our analysis indicates that a similar correlation. As demonstrated in the newly added plot for the AS system, the linear fit yields an R^2 value of 0.53. This indicates that while the general inverse relationship between ERH and viscosity persists, isolating the data by specific inorganic salts does not produce a noticeably stronger correlation compared to the overall dataset.

Revision: *"Moreover, the linear fit for organic–ammonium sulfate systems exhibits a similar correlation between ERH and viscosity, with comparable* R^2*, slope, and intercept to those obtained for the full dataset."*

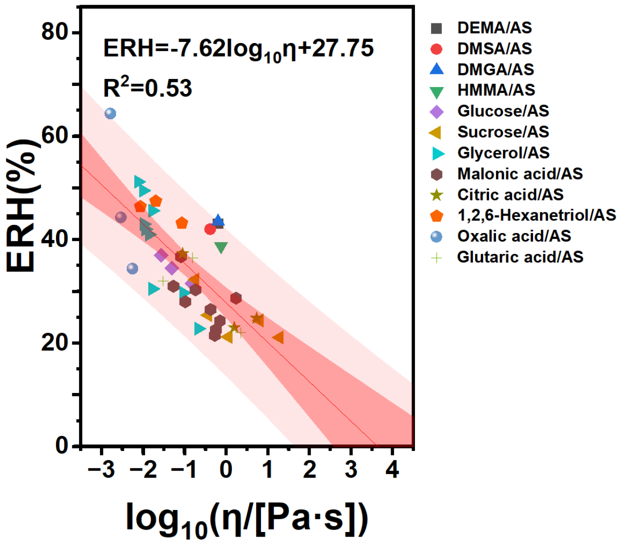

Figure S4. Linear fitting of aerosol ERH for different organic-ammonium sulfate aerosols by using viscosity ($\log_{10}\eta$*) as the predictor. Data points are color- and shape-coded to distinguish between different organic and inorganic aerosol species. The red solid line represents the linear regression fit between viscosity and ERH. The dark pink band indicates the 95% confidence interval, while the light pink band shows the prediction interval. DEMA represents Diethyl malonic acid, DMSA represents 2,2-Dimethyl succinic acid, DMGA represents 3,3-Dimethyl glutaric acid, HMMA represents DL-4-Hydroxy-3-methoxymandelic acid and DHBA represents 2,5-Dihydroxybenzoic acid.*

13. Line 180, Eq. (4): clarify the units of ERH the result from the right-hand-side of this expression. Presumably the expression yields percentage values, i.e. mathematically that is ERH*100 % on the left-hand-side and should be made clear. Also, the units of eta in the log_10 should be clarified (and for mathematical consistency normalized by the unit of eta as in the caption of Fig. 1).

Response: We thank the reviewer for pointing this out. We have clarified the units for both ERH and viscosity (η) in the revised manuscript. Following your constructive suggestion, we have updated the equation and the surrounding text to ensure strict mathematical consistency. We have explicitly indicated that the left-hand side represents ERH as a percentage, and we have normalized the viscosity η in the logarithmic term by its unit (Pa • s) to render it dimensionless.

The revised equation now reads as follows:

Revision:

$$ERH(\%) = (-10.23 \cdot \log_{10}(\eta/(Pa \cdot s)) + 27.40) \cdot 100\% \tag{6}$$

$$ERH(\eta) = \begin{cases} (-10.23 \cdot \log_{10}\eta + 27.4) \cdot 100\%, & \eta < 4.76 \times 10^2 \mathrm{Pa \cdot s} \\ 0, & \eta \geq 4.76 \times 10^2 \mathrm{Pa \cdot s} \end{cases} \tag{7}$$

$$ERH(\eta, T_g) = \left(0.25 \cdot (T_g(\omega_{org})/K) - 12.58 \cdot \log_{10}(\eta/(Pa \cdot s)) - 13.28\right) \cdot 100\% \tag{8}$$

14. Line 186: define what ERH_ing is. The definition given in the caption of Figure S1 is unclear since there is no such thing as "the ERH of the inorganic salt". Do you mean that ERH_Ing refers to the ERH of the involved salt when considering the corresponding binary aqueous salt system at the same temperature?

Response: We sincerely thank the reviewer for pointing out this imprecise terminology. We agree that the phrase “the ERH of the inorganic salt” is technically inaccurate. Your understanding is exactly what we intended. ER_{Ing} refers to the ERH of the corresponding binary aqueous inorganic salt system at the same temperature. We have revised the manuscript in both the main text and the caption of Figure S1 to clearly reflect this accurate definition and avoid any confusion.

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 (ERH/ERH_{Ing}) was even weaker, where ERH_{Ing}, represents the ERH of the corresponding pure binary aqueous inorganic salt system at the same temperature.”*

Figure S1 caption: “The ERH of the organic-inorganic mixture systems was normalized according to ERH_{Ing}, which represents the ERH of the corresponding pure binary aqueous inorganic salt system at the same temperature (i.e., ERH/ERH_{Ing}).”

15. Line 191: several typos in the sentence

Response: Thank you for pointing this out. The typos in Line 191 have been corrected.

16. Figure 3: x-axis are mislabeled between (a) and (b). For (a) should the y-axis be "Predicted ERH"? Also, the values at 0% ERH, i.e. the predicted and measured absence of ERH, should probably not be included in the correlation coefficient calculation (or provide both options). If there are a lot of points there, they mask the true predictive skill for ERH > 0.

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". We thank the reviewer for this insightful observation regarding Figure 3b. We have re-performed the fitting analysis after excluding the (0,0) data points. The revised fitting results are presented in the figure below and added to the supplementary information. The slope of the fitted curve remain close to unity and the coefficient of determination (R^2) indeed decreases. The new fitting equation is $y = 1.06x - 4.89$, $R^2 = 0.68$.

Revision: *"By performing the fitting analysis excluding the (0,0) data points (Figure S6), the slope remains close to unity, while the coefficient of determination decreases but remains significant ($R^2 = 0.68$)."*

Figure S5:

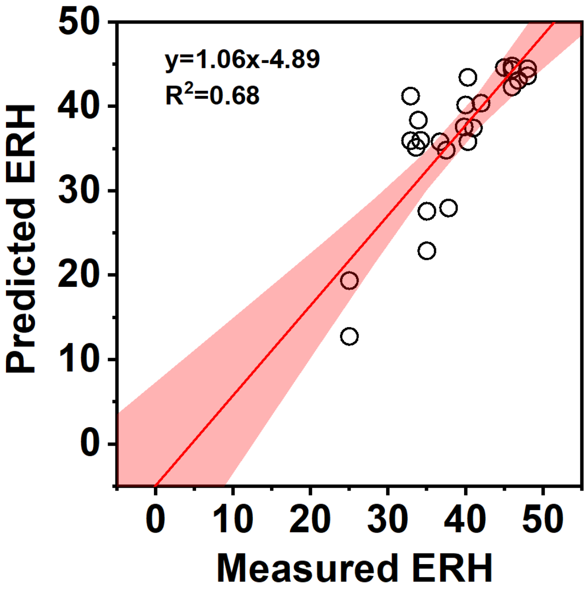

Figure S5. Comparison of measured ERH values versus model predicted ERH values based on the viscosity-ERH model by excluding the datapoints that are not effloresced (i.e., (0,0) data in Figure 3). The red line representing the linear regression fit to the data, along with the corresponding coefficient of determination (R^2). The fitted equation is $y = 1.06x - 4.89$, with slope close to unity and $R^2 = 0.68$. The pale pink shading represents the 95% confidence interval.

17. Line 246: If viscosity is not kinetically controlled, but efflorescence in supersaturated solutions is, then what is predicted through a relationship with viscosity leads to a deterministic outcome. Other methods for deterministic ERH prediction, such as that by Hodas et al. (2016) should then be discussed/considered too.

Response: We sincerely thank the reviewer for this perceptive and constructive comment, and for directing us to the significant work by Hodas et al. (2016).

In our framework, we explicitly consider thermodynamics and kinetics as complementary aspects of the same phase-transition process:(i)If an aqueous solution is not thermodynamically supersaturated with respect to the potential solid crystalline phase, efflorescence is thermodynamically prohibited, regardless of kinetic factors. As demonstrated by the method described by Hodas et al. (2016) the degree of salt supersaturation (e.g., relative to the ion activity product, IAP) defines this deterministic thermodynamic driving force. Without sufficient supersaturation, crystal formation cannot occur. (ii) Once the thermodynamic threshold is surpassed, the actual transition point is heavily modulated by the kinetic barrier to nucleation and crystal growth. Here, aerosol viscosity plays a central role. Our results indicate that viscosity serves as an effective proxy for this kinetic barrier.

To further elucidate the mechanistic basis of the model, we examined (i) viscosity changes for fixed chemical systems across RH (20–60%) and (ii) the correlation between viscosity at fixed RH and ERH. The slopes of the viscosity–RH relationships for representative systems range from −10 to −60 (Table S12), while the viscosity–ERH correlation weakens substantially when RH is held constant. These results indicate that the observed viscosity–ERH relationship primarily arises from the RH dependence of viscosity and confirm that efflorescence is governed by nucleation and crystal growth kinetics controlled by viscosity at the ERH. Accordingly, the viscosity–ERH model can be interpreted as a representative linear relationship that captures variability due to differences in organic composition and organic–inorganic interactions. It further defines a boundary condition for aerosol phase transitions: below the fitted viscosity–ERH line, aerosols tend to undergo efflorescence, whereas above this line (or beyond the threshold viscosity), aerosols tend to remain viscous without crystallization.

The corresponding discussions above have been added to the revised manuscript.

18. Line 256: use consistent notation; seemingly log here is log_10 since in Eq. (8) ln is used as well.

Response: We thank the reviewer for the suggestion. We have corrected "log" to "$\log_{10}$" in the equations to ensure consistency in notation.

19. Lines 264-268: Consider also the following: in highly viscous aerosol, the measured RH outside of a particle/sample may differ substantially from the water activity inside the droplet phase where crystallization may occur. Hence, the fact that ERH is low or efflorescence absent in experimental setups may be due to the fact that the local water content and related water activity during drying gets "locked in" and the degree of supersaturation remains lower. Hence, the question of experimental protocol followed may matter here as well. Maybe also provide an estimate of the particle equilibration timescale for the upper limit of 4.76E2 Pa s established when considering the typical particle sizes used in the experimental studies on ERH (perhaps based on the framework from Koop et al., 2011).

Response: Following your excellent suggestion, we utilized the theoretical framework outlined by Koop et al. (2011)and Shiraiwa et al. (2011) to estimate the equilibration timescale (τ) at our established upper viscosity limit of $\eta = 4.76\times10^2$ Pa·s.

First, the diffusion coefficient of water (D_{H_2O}) was estimated using the Stokes-Einstein equation ($D=kT/6\pi\eta r$). Assuming a typical hydrodynamic radius for water of $r=0.15$ nm at 298 K, we obtained $D_{H_2O}\approx3.05\times10^{-15}$ m^2/s.

Subsequently, we calculated the e-folding equilibration timescales ($\tau=d_p^2/4\pi^2D$) for two typical particle size ranges used in ERH measurements:

For submicron particles (e.g., d_p=100 nm), typical of HTDMA measurements, the equilibration timescale is extremely short (τ≈0.08 s).

For super micron particles (e.g., d_p=10μm), typical of optical microscopy measurements, the timescale increases dramatically by four orders of magnitude (τ≈830 s, or ~14 minutes).

If one considers the diffusion of larger solute ions or organic molecules, this timescale extends to well over an hour. In experiments utilizing super micron particles, typical drying rates might outpace the bulk diffusion of water when viscosity approaches 10^2-10^3 Pa·s . Consequently, water becomes kinetically trapped, maintaining a lower actual supersaturation state inside the particle and artificially delaying or completely suppressing efflorescence.

In the experiments included in our dataset, a full dehydration cycle from >80% RH to <10% RH typically lasted 60–90 minutes, with a gradual RH change (~0.067% s^{-1}) to ensure equilibrium conditions, resulting in a whole experiment period of several hours. This timescale is larger than the estimated equilibrium time for micrometer droplets and which ensures that the absence of efflorescence observed in these studies are valid under the corresponding experimental conditions. Moreover, in one of the study of (Sun. et al, 2023), an extendended observation was conducted for viscous sucrous/ammonium nitrate aerosols under dry conditions (RH: 1.8% -2.2%) for 30 hours, and no efflorescence was observed, suggesting that the observed absence of efflorescence are under equilibrium conditions.

Revision: Please referrer to our Revision in our response to your Major comment 4.

20. Line 280: correct the phrasing; the consequence is in opposite order, lower water content leads to lower water activity in the particle phase.

Response: We thank the reviewer for catching this error. We have corrected the phrasing in the revised manuscript as suggested.

Revision: *“Although SOA viscosity may significantly increase by several orders of magnitude as water content, and consequently aerosol water activity, decreases,...”*

21. Line 299: this statement is a big leap from the content of the rest of the paper. To my knowledge, whether including viscosity parameterizations in global climate models "will be essential" or negligible remains unclear.

Response: We sincerely thank the reviewer for this valuable comment. We agree that the broader implications of incorporating viscosity parameterizations into global climate models remain uncertain and require further quantitative evaluation. Accordingly, we have revised the statement to present this aspect as a future research direction rather than a definitive conclusion.

Revision: "*Further quantitative evaluation is needed to assess the extent to which viscosity-based predictive tools for aerosol phase state influence simulations of climate and air quality.*"

22. Tables S1, S2: please list the units of ERH and the temperature or the range of temperatures considered for the data listed.

Response: We thank the reviewer for the suggestion. We have updated Tables S1 and S2 in the Supplementary Information to include the units for ERH and the specific temperature conditions.

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