Chiara Giorio, Professor
Editor
Atmospheric Chemistry and Physics
University of Cambridge

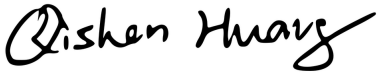
Dear. Dr. Giorio,

My co-authors and I are pleased to submit a revised version of the manuscript, titled "Predicting Organic–Inorganic Aerosol Efflorescence Using Thermodynamically Modeled Viscosity". We express our sincere gratitude to the reviewer for the insightful comments and positive assessment of our manuscript. In this revision, we have carefully responded to all reviewers' concerns, comments, and recommendations. Our responses include supplementary clarifications, additional data, supporting discussions on our key findings, and relevant references.

In this response to reviewers, we list exact comments from reviewers, followed by our point-by-point responses and specific revisions as it appears in the text. Our response and revision are in blue. Specific changes are highlighted in red in the marked manuscript, and corresponding materials are added in the updated Supporting Information. The response to each referee and the corresponding page numbers are listed as follows:

We believe that these substantial changes have resulted in a much stronger manuscript. We sincerely hope that our work aligns with your expectations, and we greatly appreciate the time and effort that you and the reviewers have invested in our work.

Sincerely,



Qishen Huang

Referee 1:

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.

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±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±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 "observaltional data" to "observational data".

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

Page 6, line 156:Change "assumted" 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.

Referee 2:

We sincerely thank the reviewer for their careful review of our manuscript and for the overall positive feedback. We highly appreciate the reviewer for highlighting the major concern regarding our methodology and the need for further analysis. We agree that viscosity is a strong function of relative humidity (RH), and the tests proposed by the reviewer have helped us clarify the underlying nature of the observed correlation. In response to this major concern, we have performed additional quantitative analyses as suggested by the reviewer. Furthermore, we have addressed all other comments, which included re-performing the fitting analysis after excluding the (0,0) data points and systematically comparing our laboratory timescales with atmospheric timescales to properly evaluate the kinetic limitations of efflorescence. Our detailed, point-by-point responses to the reviewer's comments, along with the corresponding revisions made to the manuscript, are provided below.

Major comment:

1. Line 107: The viscosity that is being correlated with the ERH is the viscosity at that ERH. Viscosity is a very strong function of RH, therefore, it is not surprising that there is a strong correlation between viscosity at the ERH and ERH. It may still be true that there is a deeper connection between viscosity and ERH, but that has not been proven without further analyses. Some analyses that could help answer this question and that I suggest include:

A. For a "representative" aerosol particle chemical system, how much does the viscosity change from say 60% RH to say 20% RH? If the change in viscosity for a fixed chemistry is comparable to the slope in Figure 2, that would suggest that most of the previously observed correlation is simply because viscosity is changing as a function of RH, rather than a deeper connection between viscosity and ERH. If the change in viscosity for a fixed chemistry is less than the slope in Figure 2, then that would suggest that viscosity being a function RH is part, but not all of the story, and that should be quantitatively discussed.

B. At a fixed RH, calculate the viscosity for each chemical system. If the correlation

between viscosity and ERH is greatly reduced, this would also suggest that most of the previously observed correlation was due to the general correlation between viscosity and RH. If the correlation largely remains, that would suggest that there is a deep connection between viscosity and ERH, which would be very exciting! If the result is somewhere in the middle, as seems perhaps most likely, then again there should be some quantitative discussion of how much of the correlation is attributable to each factor.

Response: We sincerely thank the reviewer for this insightful comment and for suggesting these constructive quantitative analyses. We agree that viscosity is a strong function of relative humidity (RH), and these proposed tests have helped us better clarify the nature of the observed correlation.

Overall, our results indicate that the reported viscosity–ERH relationship is largely influenced by the dependence of viscosity on RH, as viscosity at the ERH directly governs nucleation kinetics. An intrinsic, independent correlation between modeled viscosity and ERH is not explicitly observed. Therefore, the viscosity–ERH model established in this study should be interpreted as a representative linear relationship that captures variability arising from differences in organic composition and organic–inorganic interactions.

We further interpret the model as defining 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 and resist crystallization.

The results of the two suggested analyses are summarized below:

(a) Viscosity change for fixed chemical systems across RH:

Following your first suggestion, we calculated viscosity changes for all organic–inorganic systems in the training set over RH = 20–60%, with representative systems shown in Table R1. Linear regression of viscosity as a function of RH for fixed-composition systems yielded slopes ranging from approximately −13.20 to −45.89. Across all systems, the experimentally derived slopes range from −10 to −60.

In Figure 2, the overall fitted slope is −10.23 ($y = -10.23x + 27.4$). Since the magnitude of viscosity changes for fixed compositions is comparable to, and often

greater than, the global slope, this suggests that the observed viscosity–ERH relationship is strongly driven by the RH dependence of viscosity, while also reflecting compositional variability.

Table R1 The fitted viscosity-ERH equation for representative organic-inorganic aerosol systems with RH ranged from 20% - 60%.

Compound	OIR	20	30	40	50	60	Fitting equation
Glucose/AS	1:4	-0.13	-0.64	-1.00	-1.28	-1.56	y = -28.18x + 14.03
Oxalic acid/AS	1:3	1.64	0.63	-0.19	-0.83	-1.35	y = -13.20x + 39.74
Malonic acid/AS	1:3	-1.50	-1.80	-2.02	-2.21	-2.38	y = -45.89x - 50.99
Glutaric acid/NaCl	1:3	-1.02	-1.46	-1.74	-1.94	-2.15	y = -35.30x - 18.74
Sucrose/$NaNO_3$	1:2	-0.036	-0.74	-1.23	-1.65	-1.98	y = -20.41x + 16.96

(b) Correlation between viscosity and ERH at fixed RH:

Following your second suggestion, we evaluated the correlation between viscosity and ERH at fixed RH values (20%, 30%, 50%, and 60%) across different chemical systems. In all cases, the correlation was significantly reduced when RH was held constant.

Together, these analyses indicate that the strong correlation between viscosity at ERH and ERH itself is primarily attributable to the dependence of viscosity on RH, which in turn controls nucleation kinetics and crystal growth. Accordingly, viscosity alone cannot directly predict efflorescence without considering ambient RH.

We have revised the manuscript to clarify that the viscosity–ERH model provides a boundary relationship and threshold viscosity for aerosol phase transitions, rather than a standalone predictive variable. Specifically, below the fitted viscosity–ERH line, aerosols tend to effloresce, whereas above this line (or above the threshold viscosity), aerosols tend to remain viscous without crystallization.

Revision: *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.

Table R1 *has been added to the Supplementary Information.*

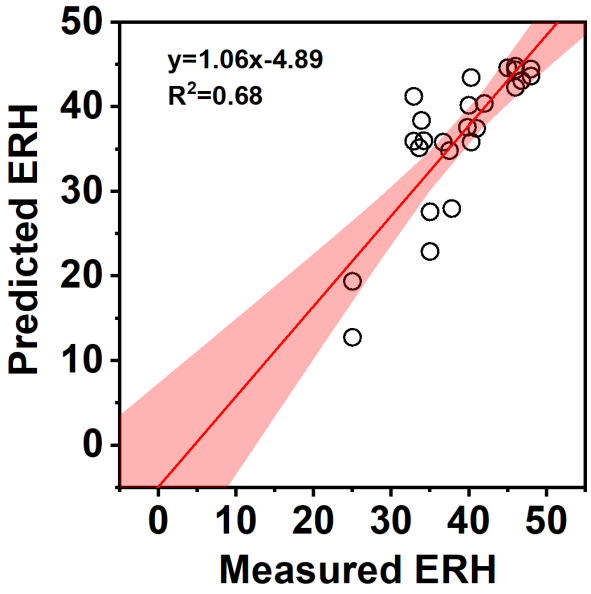
Other comments:

2. For Figure 3b, the fit is indeed close to the x=y line, but it seems like that may be partially driven by the cluster of (0,0) points. What does the fit in Figure 3b look like without those points? It seems like some of the lower viscosity points are slightly underpredicted, which is not a major concern but I think warrants at least a comment.

Response: We thank the reviewer for this insightful observation regarding Figure 3b. Following the reviewer's suggestion, we have re-performed the fitting analysis after excluding the (0,0) data points. The revised fitting results are presented in the figure below. As anticipated, the coefficient of determination (R^2) indeed decreases. The new fitting equation is $y = 1.06x - 4.89$, $R^2 = 0.68$, which aligns well with the y=x line.

We acknowledge the reviewer's observation that the ERH values for a subset of the data are underestimated by the model. We attribute this discrepancy to a potential imbalance in how the model weights the kinetic inhibition mechanisms. As relative humidity decreases, the increasing viscosity of organic-inorganic mixtures imposes kinetic limitations on crystal nucleation by restricting molecular diffusion. However, under low-RH conditions, the current model may overemphasize the inhibitory effect

of viscosity on the nucleation process when employing viscosity as a key predictive parameter.



3. If the major finding holds that viscosity controls ERH, and that high viscosity particles do not effloresce, could that mean that experimentalists are just waiting long enough to observe the efflorescence? It is worth commenting on what the typical timescale of these laboratory experimental observations are, what the typical time scales in the atmosphere are, and how long based on some simple physical principles efflorescence might be delayed for some of these higher viscosity compounds.

Response: We sincerely thank the reviewer for raising this highly insightful question regarding the kinetic limitations of efflorescence. We agree that it is crucial to clarify whether the lack of observed efflorescence in high-viscosity particles is merely an artifact of a limited experimental time window.

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 our tested dataset, the majority of the experimental data were obtained from our laboratory. A complete dehydration cycle, from high relative humidity (> 80% RH) to low relative humidity (< 10% RH), typically lasts about 60–90 minutes, with the total observation time for each experiment being several hours. When examining the physical principles of delayed efflorescence, we found a strong dependence on the

organic-to-inorganic ratio (OIR), which directly influences viscosity. For pure ammonium sulfate particles, once the critical RH range is reached, the nucleation induction time at the efflorescence point is approximately 83 s. For sucrose/ammonium sulfate mixed particles with an OIR ≤ 1:2, this induction time increases significantly to 2177 s. Furthermore, for mixtures with an OIR > 1:1, no crystallization was observed even after prolonged exposure to low RH conditions (< 10% RH). This demonstrates that increased viscosity drastically hinders molecular diffusion, thereby severely delaying or entirely inhibiting the crystal nucleation process(Wang et al., 2017).

Regarding atmospheric timescales, Anon et al. (Anon, 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 the data from Wang et al.(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, while laboratory experiments are inherently constrained by practical observation windows, both the fundamental kinetic principles (nucleation induction times) and existing literature strongly suggest that the inhibition of efflorescence in highly viscous particles holds true even under realistic atmospheric conditions.

References

Anon: Formation and cycling of aerosols in the global troposphere, Atmospheric Environment, 34, 4215–4240, https://doi.org/10.1016/S1352-2310(00)00239-9, 2000.

Wang, L.-N., Cai, C., and Zhang, Y.-H.: Kinetically Determined Hygroscopicity and Efflorescence of Sucrose-Ammonium Sulfate Aerosol Droplets under Lower Relative Humidity, J Phys Chem B, 121, 8551–8557, https://doi.org/10.1021/acs.jpcb.7b05551, 2017.

Community Comment 1:

Dear Dr. Armeli, Dr. Peters, and Dr. Koop,

We sincerely appreciate your detailed comments, which helped us identify errors and improve our manuscript. We have carefully gone through your comments and made the corresponding corrections and revisions, which are included in this document along with our explanations and responses. All revisions mentioned in this response will appear in future versions of both the main text and the SI. We hope that our manuscript has now become stronger, and we would greatly appreciate any further suggestions for improvement. Please find our detailed responses and revisions below.

Major Comment:

Response: We very much appreciate your comment correcting the Tg values presented in the original Table S4 and thank you for providing detailed explanations, including the correct input parameters, which helped us reexamine our previous calculations. After carefully reviewing our earlier calculations, we found that the Tg values were miscalculated due to the use of Tm in units of degrees Celsius instead of Kelvin, which led to deviations from the reference values.

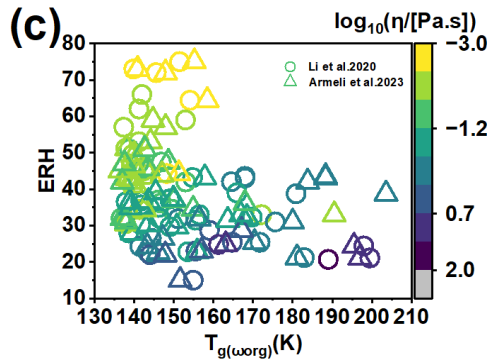
We have now corrected all T_g values using the correct Functional Group Mode (FGM) and SMILES Mode (SM), both with and without the optional Tm parameter, following your detailed instructions. Our recalculated results agree with the values provided in your comment and with those reported in your previous work. Accordingly, we have updated the manuscript in the following aspects: (1) T_g values were recalculated using the ML method from Armeli et al., 2023, and all parameters and calculated results are included in Figure 1c, S1, S2, and Tables S4 – S7; (2) Discussions about the model in the manuscript were revised to reflect its accuracy; (3) References relevant to your comments are now cited in the manuscript.

With the corrected T_g values, we show that the relationship between T_g and ERH remains weak (Figure 1c), similar to the T_g values interpolated using the parameterization from Li et al. Our main finding therefore remains robust: thermodynamically modeled viscosity continues to show a significantly stronger

correlation with ERH than T_g, and the inclusion of T_g provides only negligible statistical improvement to the viscosity-based model.

Revisions:

(1) Figure 1c now includes T_g values achieved from both Li et al. and Armeli et al.



(2) 2.3 Calculation of the fractional glass transition temperature ($Tg_{(\omega org)}$)

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., 2020b).

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 – S6.

The volatility-based parameterization developed by Li et al. (Eq. 1) predicts T_g from the saturation mass concentration (C^0).

$$T_g = 288.7 - 15.33 \times \log_{10}(C^0) - 0.33 \times [\log_{10}(C^0)]^2 \quad (1)$$

C^0 was estimated as a function of C, O, N, S numbers of the organic compound ($\log_{10}C^0 = f(n_C, n_O, n_N, n_S)$, Eq. 2): (Donahue et al., 2011; Li et al., 2016)

$$\log_{10} C^0 = (n_C^0 - n_C)b_C - n_O b_O - 2\frac{n_C n_O}{n_C + n_O} b_{CO} - n_N b_N - n_S b_S. \quad (2)$$

Here, n_C^0 is the reference carbon number; n_C, n_O, n_N, and n_S denote the number of carbon, oxygen, nitrogen, and sulfur atoms, respectively; b coefficients (b_C, b_O, b_N, and b_S) denote atomic contributions fitted via multi-linear least squares analysis from 30,000 compounds across multiple classes(CH, CHO, CHN, CHON, CHOS, and CHONS)(Li et al., 2016). $b_{(CO)}$ represents the carbon-oxygen nonideality.

A comparison between the experimentally measured T_g values of the organic compounds and the predictions derived from the methods of both Li et al.(Li et al., 2020a) and Armeli et al.(Armeli et al., 2023) is presented in Table S6.

The presence of salts inorganic salts (e.g., sodium nitrate) have been demonstrated to modulate the T_g of organic–inorganic mixtures; specifically, depending on the effective T_g of inorganic salt, the T_g of the mixture can be either elevated or depressed (Dette and Koop, 2015b). Typically, the T_g of organic–inorganic mixtures is calculated using the Gordon–Taylor equation (Gordon and Taylor, 1952) :

$$T_g = \frac{\omega_1 T_{g1} + \frac{1}{k}\omega_2 T_{g2}}{\omega_1 + \frac{1}{k}\omega_2} \quad (3)$$

where T_g represents the glass transition temperature of the binary mixture, T_{g1} and T_{g2} are the T_g values of the respective pure compounds, ω_1 and ω_2 denote the mass fractions of the two components, and k is the Gordon–Taylor constant. However, the glass transition temperature of inorganic compounds is limited, we thus employ the glass transition temperature of the organic-water system ($T_{g,org\text{-}water}$) as a proxy for the overall mixture T_g when analyzing the correlation between efflorescence relative humidity (ERH) and T_g. In Eq.(3), the parameters ω_2 and T_{g2} are substituted with the mass fraction ω_{org} and the glass transition temperature $T_{g(\omega_{org})}$ of the organic component, respectively，where ω_{org} is the organic mas fraction calculated via the AIOMFAC model at aerosol ERH. $T_{g,w}$ is the glass transition temperature of pure water (136 K) (Kohl et al., 2005), while k_{GT} (the Gordon–Taylor constant) is assumed to be 2.5 for organic–water mixtures (Koop et al., 2011; Zobrist et al., 2008). These two values are substituted for T_{g1} and k in the equation, with ω_1 calculated as (1-ω_{org}). As reported by Koop et al.

(Dette and Koop, 2015a), the T_g of $NaNO_3$ is specified as 290 K. We utilized this value to calculate the T_g for glycerol/$NaNO_3$ and sucrose/ $NaNO_3$ mixtures across a range of mixing ratios (Figure S1).

(3) Table S4 – S7: Calculation parameters and updated T_g values predicted using the ML-chemical structure method.

Table S4. Input parameters employed for the calculation of glass transition temperatures T_g using the model developed by Armeli et al.

Compound	**FG Mode**								**Optional***	**SMILES Mode**
	$-CH_3$	$-CH_2$	-CH	-C-	-OH	-O-	=O	DBE	T_m / K	SMILES String
glucose	0	1	4	1	5	0	1	1	420	C(C1C(C(C(C(O1)O)O)O)O)O)O
sucrose	0	3	8	1	8	3	0	2	457	C(C1C(C(C(C(O1)OC2(C(C(C(O2)CO)O)O)CO)O)O)O)O
glycerol	0	2	1	0	3	0	0	0	291	OCC(O)CO
citric acid	0	2	0	4	4	0	3	3	432	C(C(=O)O)C(CC(=O)O)(C(=O)O)O
1,2,6-hexanetriol	0	5	1	0	3	0	0	0	300	C(CCO)CC(CO)O
Malonic acid	0	1	0	2	2	0	2	2	408	C(C(=O)O)C(=O)O
Oxalic acid	0	0	0	2	2	0	2	0	462.5	C(=O)(C(=O)O)O
Glutaric acid	0	3	0	2	2	0	2	2	371	C(CC(=O)O)CC(=O)O
Maleic acid	0	0	2	2	2	0	2	3	403.5	C(=C\C(=O)O)\C(=O)O
DEMA	2	2	0	3	2	0	2	2	395	CCC(CC)(C(=O)O)C(=O)O
DMSA	2	1	0	3	2	0	2	2	413.5	CC(C)(CC(=O)O)C(=O)O
DMGA	2	2	0	3	2	0	2	2	376.5	CC(C)(CC(=O)O)CC(=O)O
HMMA	1	0	4	4	3	1	1	5	405	COC1=C(C=CC(=C1)C(C(=O)O)O)O

* T_m is an optional input parameter for both modes

Table S5. T_g of pure organic compounds calculated using the method of Armeli et al.

Compound	T_g(FGM with T_m))	T_g (FGM no T_m)	T_g (SM with T_m)	T_g (SM no T_m)
Glucose ($C_6H_{12}O_6$)	295.8	300.2	275.3	282.3
Sucrose ($C_{12}H_{22}O_{11}$)	344	342.4	342.5	339.1
Glycerol ($C_3H_8O_3$)	187.4	187.5	187.4	187.7
Malonic acid $C_3H_4O_4$	258.9	231.3	259	234.9
Citric acid $C_6H_8O_7$	284.4	284.4	283.6	283.2
1,2,6-Hexanetriol $C_6H_{14}O_3$	202.3	201.5	203.6	202.7
Oxalic acid $C_2H_2O_4$	276	226.8	271.9	224.9
Glutaric acid $C_5H_8O_4$	261.9	242.4	250.1	252.5
DEMA Diethyl malonic acid	274.8	266.2	271.1	270.5
DMSA 2,2-Dimethyl succinic acid	282	262.5	277.9	269.5
DMGA 3,3-Dimethyl glutaric acid	267.5	266.2	265.6	261.5
HMMA DL-4-Hydroxy-3-methoxymandelic acid	276.1	296.2	276.1	284
Maleic acid $C_4H_4O_4$	264.1	242.6	261.6	251.8

Table S6. T_g of pure organic compounds calculated using the method of Armeli et al., and the corresponding T_g of the mixed systems derived via the Gordon–Taylor equation.

Compound	ω_{org}	T_g(FGM with T_m))	T_g (FGM no T_m)	T_g (SM with T_m)	T_g (SM no T_m)
Glucose	0.19	149.74	150.12	147.98	148.58
($C_6H_{12}O_6$)/AS	0.24	154.27	154.88	152.04	152.84
	0.33	162.55	163.61	159.48	160.64
Sucrose	0.32	168.70	168.45	168.47	167.93
($C_{12}H_{22}O_{11}$)/AS	0.50	195.96	195.50	195.53	194.55
	0.33	170.63	170.37	170.38	169.82
	0.41	181.45	181.10	181.12	180.38
	0.51	197.45	196.98	197.01	196.01
Glycerol	0.10	138.12	138.12	138.12	138.13
($C_3H_8O_3$)/AS	0.18	140.03	140.04	140.03	140.06
	0.30	143.48	143.50	143.48	143.53
	0.50	150.73	150.76	150.73	150.81
	0.61	155.86	155.89	155.86	155.97
	0.09	138.05	138.06	138.05	138.06
	0.19	140.34	140.34	140.34	140.36
	0.13	138.99	138.99	138.99	139.00
	0.10	138.29	138.29	138.29	138.30
Malonic acid	0.18	145.86	143.65	145.87	143.94
$C_3H_4O_4$/AS	0.25	150.51	147.25	150.52	147.67
	0.41	162.36	156.44	162.38	157.21
	0.55	176.16	167.14	176.20	168.32
	0.23	149.00	146.08	149.01	146.46
	0.09	140.53	139.52	140.54	139.65
	0.05	138.61	138.03	138.62	138.10
	0.17	145.43	143.31	145.43	143.59
	0.18	145.84	143.63	145.84	143.91
Citric acid	0.21	149.89	149.89	149.82	149.78
$C_6H_8O_7$/AS	0.36	163.11	163.11	162.97	162.89
	0.29	157.00	157.00	156.88	156.83
1,2,6-Hexanetriol	0.37	148.78	148.62	149.03	148.86
$C_6H_{14}O_3$/AS	0.13	139.91	139.86	139.98	139.93
	0.56	158.11	157.84	158.54	158.24
Oxalic acid	0.34	159.55	151.28	158.86	150.96
$C_2H_2O_4$/AS	0.45	170.60	158.44	169.59	157.97
	0.17	146.36	142.72	146.06	142.58
Glutaric acid	0.24	150.04	147.86	148.72	148.99
$C_5H_8O_4$/AS	0.02	137.14	136.96	137.03	137.05
	0.07	139.41	138.88	139.09	139.16
DEMA/AS	0.63	191.78	188.32	190.29	190.05

Diethyl malonic acid DMSA/AS		191.16	183.79	189.61	186.44
2,2-Dimethyl succinic acid DMGA/AS	0.60	188.84	188.32	188.08	186.43
3,3-Dimethyl glutaric acid HMMA/AS	0.63	195.10	203.58	195.10	198.44
DL-4-Hydroxy-3-methoxymandelic acid	0.65				
Oxalic acid/KCl	0.12	143.47	140.85	143.25	140.74
$C_2H_2O_4$	0.27	154.35	147.90	153.81	147.65
	0.40	165.60	155.20	164.73	154.80
Malonic acid/KCl	0.19	146.47	144.12	146.48	144.42
$C_3H_4O_4$	0.03	137.31	137.02	137.31	137.05
Maleic acid/KCl	0.22	149.26	147.04	149.00	147.99
$C_4H_4O_4$	0.51	173.15	166.92	172.43	169.59
	0.72	201.50	190.51	200.22	195.21
Glutaric acid/NaCl	0.18	146.34	144.74	145.37	145.57
$C_5H_8O_4$	0.01	136.44	136.37	136.40	136.41
	0.20	147.14	145.42	146.10	146.31
	0.64	188.06	180.00	183.18	184.18
	0.75	204.92	194.24	198.46	199.77
Malonic acid /NaCl	0.56	177.21	167.95	177.24	169.16
$C_3H_4O_4$	0.63	186.36	175.05	186.40	176.52
Glycerol /$NaNO_3$	0.43	147.91	147.93	147.91	147.98
($C_3H_8O_3$)	0.27	142.62	142.63	142.62	142.65
	0.07	137.55	137.55	137.55	137.56
	0.14	139.11	139.12	139.11	139.13
	0.52	151.57	151.60	151.57	151.66
	0.42	147.54	147.56	147.54	147.60
	0.64	157.50	157.54	157.50	157.63
Sucrose/$NaNO_3$	0.45	187.29	314.83	183.84	186.08
($C_{12}H_{22}O_{11}$)	0.28	163.85	307.44	161.98	163.20

Table S7. Comparison of the experimentally measured T_g of organic compounds with values predicted using the method of Li et al. and the method of Armeli et al.

	Tg / K					
Compound	**$T_{g\ literature}$(Armeli Iapichino et al., 2023)**	**$T_{g\ Li}$(Li et al., 2020)**	**$T_{g\ Armeli}$(Armeli et al., 2023)**			
			FGM with T_m	FGM no T_m	SM with T_m	SM no T_m
Glucose	303	253.32	295.8	300.2	275.3	282.3
Sucrose	334	350.97	344.0	342.4	342.5	339.1
Glycerol	186	181.38	187.4	187.5	187.4	187.7
Citric acid	284.35	274.53	284.4	284.4	283.6	283.2
1,2,6-Hexanetriol	204.15	192.25	202.3	201.5	203.6	202.7
Mean Absolute Error / K		18.6	4.0.	3.1	7.8	6.0

(4) Figure S1 & S2: T_g of organic-$NaNO_3$ mixture and the comparison between the use of different T_g predictions.

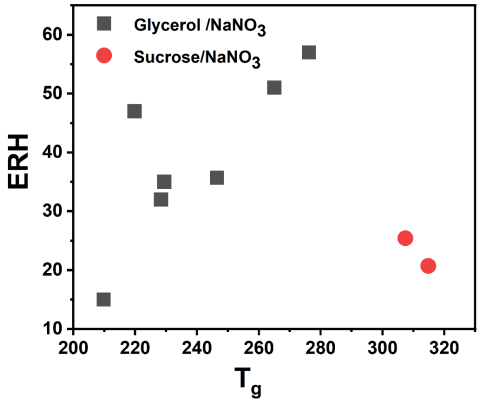


Figure S1. Calculated T_g of glycerol/$NaNO_3$ and sucrose/$NaNO_3$ systems at various mixing ratios, plotted against their respective efflorescence points. The calculations are based on the reported Tg of 290 K for $NaNO_3$.

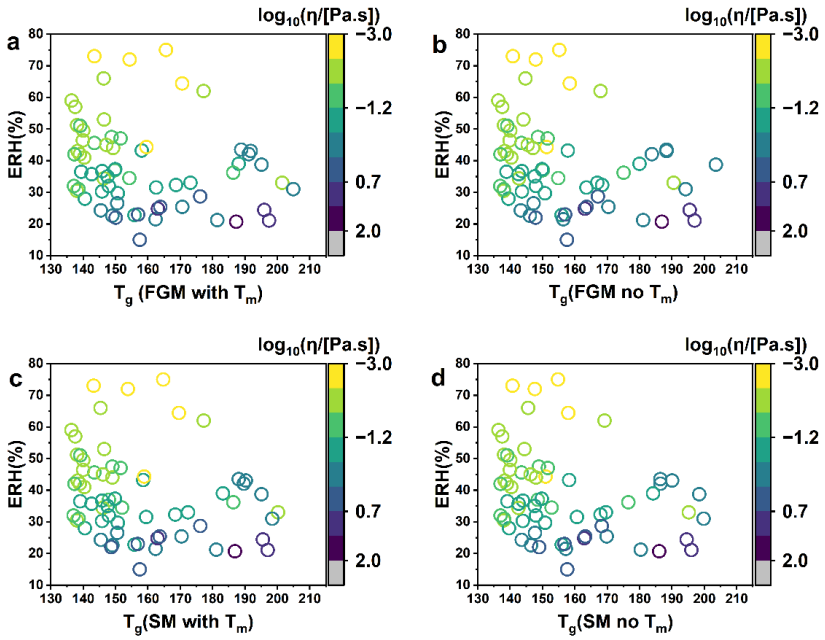


Figure S2. Relationships between the ERH of organic-inorganic mixed aerosols. (a) T_g(FGM with T_m), (b) T_g(FGM no T_m), (c) T_g(SM with T_m), and (d) T_g(SM no T_m), T_g of organic compounds calculated using the methodology and online predictor of Armeli et al. (Armeli et al., 2023), and the resulting T_g of organic–inorganic mixtures derived via the Gordon–Taylor equation.

Additional comments

（1）On page 6 of the main text the authors state "Since inorganic salts involved in this study do not exhibit T_g due to crystallization […]". However, even if T_g of pure inorganic salts such as sodium nitrate cannot be measured directly due to crystallization, such salts have been shown to influence the T_g of organic/inorganic mixtures. For example, depending upon the T_g of the pure organic, inorganic salts have shown to increase or decrease the T_g of the mixture (cf. Figure 2 and 4 in Dette and Koop, 2015). Therefore, it appears to us that ignoring the effect of the inorganic salt on T_g by representing it only by the fractional T_g of the organic phase may lead to a misrepresentation. In some cases, for example the binary mixture of sodium nitrate and sucrose, the T_g of the mixture over nearly the entire composition range is known and could be used instead, or at least used as a test case for comparison.

Response: We sincerely thank the reviewer for this insightful comment and for directing our attention to the important work of Dette and Koop (Dette and Koop, 2015a). We entirely agree with your point: inorganic salts can indeed significantly

influence the glass transition temperature T_g of organic/inorganic mixtures. Our initial decision to simplify the calculation by representing the mixture's T_g primarily through the organic phase was fundamentally constrained by the limited availability of reliable, experimentally derived T_g data for pure inorganic salts and their specific multi-component mixtures across our broader dataset. We therefore decided to use the fractional T_g values for the organic/water mixtures which are also derived from the Gordon–Taylor equation, which ensures that the underlying mathematical basis remains the same. To address your concern and test whether this proxy method leads to a misrepresentation of our findings, we followed your suggestion and conducted a supplementary correlation analysis. We calculated the T_g of the mixtures by incorporating the T_g of pure $NaNO_3$. The results of this re-evaluation demonstrate that, regardless of whether the pure inorganic salt T_g or water T_g is used in the Gordon-Taylor framework, the newly calculated T_g of the organic/inorganic mixtures still exhibits no apparent correlation with the ERH. Therefore, while the inclusion of the inorganic component's T_g should be more accurate and shifts the T_g values, it does not alter the fundamental relationships observed, nor does it affect the major findings of this study. We have revised the main text to acknowledge the influence of inorganic salts on mixture T_g ((Dette and Koop, 2015b)), explained why our core conclusion remains robust despite the data constraints. We deeply appreciate this crucial discussion, and look forward to further investigating the complex role of inorganic salt T_g in mixed systems. We believe that more experimental T_g data of pure inorganic salts are important in understanding aerosol physiochemical behaviors.

Revisions:

(1) Typically, the T_g of organic–inorganic mixtures is calculated using the Gordon–Taylor equation (Gordon and Taylor, 1952) :

$$T_g = \frac{\omega_1 T_{g1} + \frac{1}{k}\omega_2 T_{g2}}{\omega_1 + \frac{1}{k}\omega_2} \quad (3)$$

where T_g represents the glass transition temperature of the binary mixture, T_{g1} and T_{g2} are the T_g values of the respective pure compounds, ω_1 and ω_2 denote the mass fractions

of the two components, and k is the Gordon–Taylor constant. However, the glass transition temperature of inorganic compounds is limited, we thus employ the glass transition temperature of the organic-water system ($T_{g,org\text{-}water}$) as a proxy for the overall mixture T_g when analyzing the correlation between efflorescence relative humidity (ERH) and T_g. In Eq.(3), the parameters ω_2 and T_{g2} are substituted with the mass fraction ω_{org} and the glass transition temperature $T_{g(\omega_{org})}$ of the organic component, respectively，where ω_{org} is the organic mas fraction calculated via the AIOMFAC model at aerosol ERH. $T_{g,w}$ is the glass transition temperature of pure water (136 K) (Kohl et al., 2005), while k_{GT}(the Gordon–Taylor constant) is assumed to be 2.5 for organic–water mixtures (Koop et al., 2011; Zobrist et al., 2008). These two values are substituted for T_{g1} and k in the equation, with ω_1 calculated as (1-ω_{org}). As reported by Koop et al. (Dette and Koop, 2015a), the T_g of $NaNO_3$ is specified as 290 K. We utilized this value to calculate the T_g for glycerol/$NaNO_3$ and sucrose/ $NaNO_3$ mixtures across a range of mixing ratios (Figure S1).

(2) As indicated in Table S6, the Functional Group Mode without T_m (FGM without T_m) exhibits the minimum Mean Absolute Error. Consequently, this specific mode was selected to calculate the T_g of the mixtures for a comparative analysis with the parameterization developed by Li et al. (Li et al., 2020a). 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 S1). Similarly, no discernible correlation with ERH was observed.

（2）It is unclear to us why the authors used a training set and a test set because, as far as we understand, they perform a least-squares regression. This splitting procedure is followed in ML algorithms to avoid overfitting. However, in our opinion, overfitting in least-squares regression can be avoided by using a simple regression formula with not too many parameters, as it is done in the current procedure of Chen et al. 2026[1] . If the authors prefer to use a training set and a test set, would a cross-validation procedure be advantageous when splitting up the dataset, as it would allow for a more

robust assessment of the model's generalizability than a single training-test split?

Response: We sincerely thank the reviewer for raising this point. We completely agree that for a simple least-squares regression with few parameters, overfitting is generally not a primary concern. Here, we would like to clarify the rationale behind our dataset division. Our splitting procedure was a separation based on the source of the 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 research group, 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. We borrowed the term of training set and validation set from machine learning methods which likely causes the confusion.

Revision:

2.1 The dataset for the parameterization

…A total of 102 ERH data points for organic-inorganic mixture aerosols were collected: 66 ERH points for the training set (Table S1) and the remaining 36 points for the validation set (Table S2). The training set includes aerosol compositions containing 14 organic compounds (composed of C, H, and O) and 4 inorganic salts (ammonium sulfate, ammonium nitrate, sodium chloride, and potassium chloride) mixed in varying ratios. All ERH data in the training were from our previous studies, and all corresponding viscosity data were derived using the AIOMFAC model. The validation set contains 36 ERH data from systems involving 12 organic compounds and 4 inorganic salts (Table S2), selected from studies that either reported both aerosol ERH and viscosity reported or observed no efflorescence…

(3) A comparison of Figures 2 and S1 reveals that the Glucose/AS mixture is listed twice in the legend of Figure 2, and the Sucrose/$NaNO_3$ mixture is missing.

Response: We thank the reviewers for their careful inspection of the figures. In the revised manuscript, we have corrected the legend to ensure it is fully consistent with the training dataset listed in Table S1.

Revision:

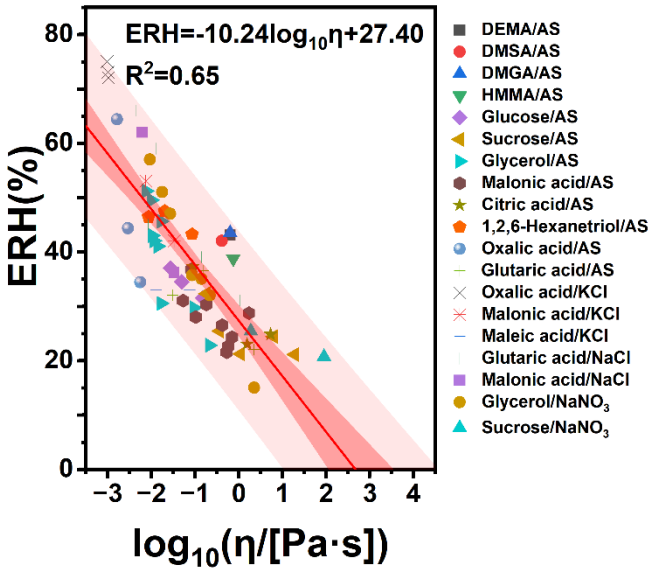


Figure 2. Linear fitting of aerosol ERH for different organic-inorganic aerosols by using viscosity ($\log_{10}\eta$) as the predictor. Data points are color- and shape-coded to distinguish between different organic and inorganic aerosol species. The red solid line represents the linear regression fit between viscosity and ERH. The dark pink band indicates the 95% confidence interval, while the light pink band shows the prediction interval. AS refers to ammonium sulfate, 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.

(4) In equation (5) and Figure 2, the authors report a threshold viscosity of 4.76×10^2 Pa□s. Could they also provide the 95% confidence intervals and the prediction intervals for this value?

Response: We have provided the 95% prediction interval for the threshold viscosity. Based on the regression analysis, the threshold viscosity is determined to be 4.76×10^2 Pa·s, and the corresponding 95% confidence interval is [1.05×10^2, 3.24×10^3] Pa·s. We

have updated the discussion about Eq. (5) to include these values.

(5) In lines 139−140, the authors state that our T_g model is a Tm-based method. This statement is incorrect because our ML model is a chemical structure-based model and does not require information of Tm . However, it allows Tm to be included as an additional input parameter, which slightly improves its predictions (see Table A for comparison).

Response: We sincerely appreciate your kind reminder for the mischaracterization of your model in our manuscript. We have corrected the text in the revised manuscript to accurately describe your machine-learning model as a chemical structure-based method rather than a T_m-based one. We have also explicitly clarified that while T_m is not a mandatory input, including it as an optional parameter can further enhance the model's predictive accuracy. The revised paragraph now properly distinguishes between volatility-based, T_m-based, and chemical structure-based parameterizations.

Revision: Have been included in "2.3 Calculation of the fractional glass transition temperature ($Tg_{(\omega org)}$)" above.

The references below are now cited in the revised manuscript.

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.

Dette, H. P. and Koop, T.: Glass Formation Processes in Mixed Inorganic/Organic Aerosol Particles, J. Phys. Chem. A, 119, 4552–4561, https://doi.org/10.1021/jp5106967, 2015a.

Dette, H. P. and Koop, T.: Glass Formation Processes in Mixed Inorganic/Organic Aerosol Particles, J. Phys. Chem. A, 119, 4552–4561, https://doi.org/10.1021/jp5106967, 2015b.

Referee 3:

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.

1. Page 6 line 155: Why kGT is assumed to be 2.5 for all organic water mixtures. There are references provided, however a sentence or two justifying this assumption is good.

Response: We thank the reviewer for this insightful comment. The assumption of k_{GT} = 2.5 for all organic–water mixtures is an estimate based on established practices in the literature for secondary organic aerosol components. While the exact k_{GT} value depends on specific solute–water interactions, and can be precisely determined for well-investigated individual systems (e.g., k_{GT} =3.18 for citric acid), obtaining compound-specific parameters for every constituent within highly complex SOA mixtures presents practical challenges. In the absence of explicit experimental data for all constituent compounds, k_{GT} =2.5 is widely utilized as a robust and representative baseline derived from structurally related water-soluble organics. We have explicitly justified this assumption in the revised manuscript and acknowledged the associated uncertainty (typically evaluated as ±1.0) 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±1.0). This variability introduces a potential, albeit minor, source of uncertainty into the T_g-based model predictions presented in this study...*

2. *What is the pH of these aerosol systems? pH impacts on aerosol properties greatly, and thus authors may want to comment on that.*

Response: We thank the reviewer for raising this important question. We agree that

aerosol pH is a critical parameter that can influence aerosol phase transitions. In our experiments, the aerosol droplet pH ranged from approximately 3.0 to 5.0, which is consistent with the typical pH range observed for tropospheric aerosols. One of our previous studies examined the impact of pH on aerosol efflorescence (Sun et al., 2023); which is one of the few studies that reports the effect of pH on aerosol efflorescence. However, to the best of our knowledge, no consistent trend has been established regarding the direction or magnitude of pH effects on ERH, since the influence of pH is highly dependent on the specific organic composition. In the present study, we consider the influence of pH to be implicitly included within the overall model uncertainty. We have now added a brief discussion in the revised manuscript to acknowledge and contextualize the potential role of pH in modulating ERH.

Revision: *We also note that other parameters, such as aerosol pH, can affect the ERH of organic/inorganic aerosol (Sun et al. 2023); however, to the best of our knowledge, no consistent trend has been established regarding the direction or magnitude of this effect, since the influence of pH is highly dependent on the specific organic composition. Given that the modeled viscosity accounts for organic functional groups, the influence of pH may be implicitly reflected within the overall model uncertainty. Future studies should incorporate more inorganic T_g data and systematically investigate pH effects to provide more accurate $T_g(\omega_{org})$ and pH values and better evaluate the role of both parameters in the viscosity-ERH relationship.*

2. Page 3, 10: some typos found. Please do an additional round of copy-editing.

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.

4. *Page 4, lines 100-105: Is it possible to add these equations as well to the equation lists for easy following?*

Response: We thank the reviewer for this constructive suggestion. We agree that explicitly formatting these descriptions as numbered equations greatly improves the readability and flow of the manuscript. Accordingly, we have added the requested

equations corresponding to the text on Page 4, lines 100-105, and updated the equation numbering sequentially throughout the revised manuscript.

Revision:

$$O{:}C_{mixture} = \left(\frac{n_{O,org}}{n_{C,org}}\right) \cdot x_{org} \quad (1)$$

$$x_{org} = \frac{n_{org}}{n_{org} + n_{inorg}} \quad (2)$$

5. Please expand the caption 3 to identify all abbreviations in the figure.

Response: We thank the reviewer for the careful check. As suggested, we have expanded the caption of Figure 3 to explicitly define all abbreviations used in the figure, ensuring better clarity for the readers.

Revision:

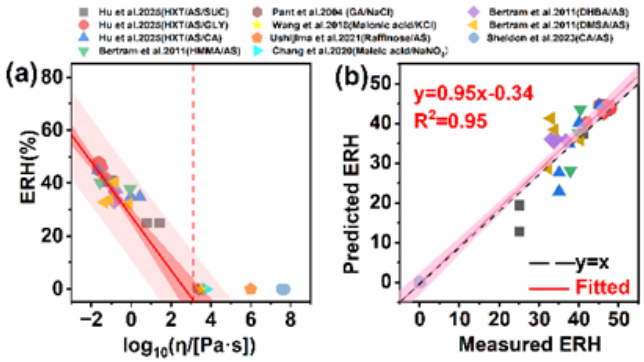


Figure 3. (a) Comparison between the validation set data and viscosity-ERH model prediction (Eq. 4). (b) Comparison of measured ERH values versus model predicted ERH values based on the viscosity-ERH model. The black dashed line denotes the y=x reference line, and the red line representing the linear regression fit to the data, along with the corresponding coefficient of determination (R^2), R^2=0.95. The pale pink shading represents the 95% confidence 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, CA represents citric acid, SUC represents sucrose, GA represents glutaric acid, HXT represents 1,2,6-hexanetriol.

6. Is the change in viscosity due to differences in RH (e.g., 20% vs 75%) different between chemical systems studied? If so, do they follow any relationship with the structure/ functional groups present?

Response: We sincerely thank the reviewer for raising this insightful question. The change in viscosity due to differences in relative humidity (RH) varies significantly between the different chemical systems studied. In our quantitative analysis, we

calculated the viscosity changes for all organic-inorganic aerosol systems across an RH range from 20% to 60%, and several representative systems were shown in Table R1. By performing linear regressions of viscosity as a function of RH for these fixed chemical systems, as shown in the table, we found that the slopes of the fitting equations vary considerably, ranging from approximately -13.20 to -45.89. No specific correlation between the fitted slope and functional groups were found.

Table R1 The fitted viscosity-ERH equation for representative organic-inorganic aerosol systems with RH ranged from 20% - 60%.

Compound	OIR	20%	30%	40%	50%	60%	Fitting equation[a]
Glucose/AS	1:4	-0.13	-0.64	-1.00	-1.28	-1.56	y = -28.18x + 14.03
Oxalic acid/AS	1:3	1.64	0.63	-0.19	-0.83	-1.35	y = -13.20x + 39.74
Malonic acid/AS	1:3	-1.50	-1.80	-2.02	-2.21	-2.38	y = -45.89x - 50.99
Glutaric acid/NaCl	1:3	-1.02	-1.46	-1.74	-1.94	-2.15	y = -35.30x - 18.74
Sucrose/$NaNO_3$	1:2	-0.036	-0.74	-1.23	-1.65	-1.98	y = -20.41x + 16.96

[a] y denotes aerosol ERH and x denotes the $\log_{10}\eta$ at corresponding RH values.

Referee 4:

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 $Tg(\omega_{org})$, 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 R², slope, and intercept to those obtained for the full dataset."*

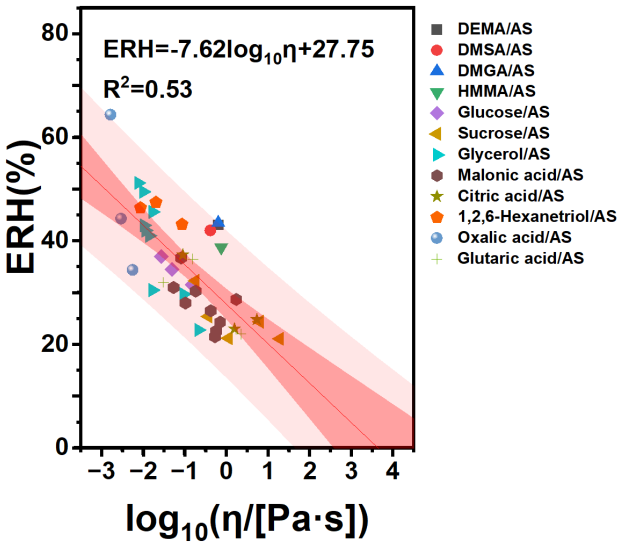


Figure S4. Linear fitting of aerosol ERH for different organic-ammonium sulfate aerosols by using viscosity ($\log_{10}\eta$) as the predictor. Data points are color- and shape-coded to distinguish between different organic and inorganic aerosol species. The red solid line represents the linear regression 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:

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

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

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

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:

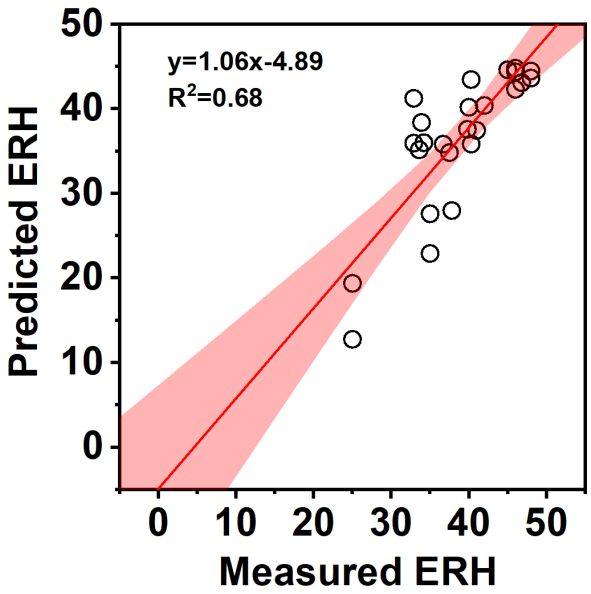


Figure S5. Comparison of measured ERH values versus model predicted ERH values based on the viscosity-ERH model by excluding the datapoints that are not effloresced (i.e., (0,0) data in Figure 3). The red line representing the linear regression fit to the data, along with the corresponding coefficient 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 ($\tau\approx0.08$ 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.

References

Anon: Formation and cycling of aerosols in the global troposphere, Atmospheric Environment, 34, 4215–4240, https://doi.org/10.1016/S1352-2310(00)00239-9, 2000.

Bouzidi, H., Fayad, L., Coeur, C., Houzel, N., Petitprez, D., Faccinetto, A., Wu, J., Tomas, A., Ondráček, J., Schwarz, J., Ždímal, V., and Zuend, A.: Hygroscopic growth and CCN activity of secondary organic aerosol produced from dark ozonolysis of γ-terpinene, Science of The Total Environment, 817, 153010, https://doi.org/10.1016/j.scitotenv.2022.153010, 2022.

Ciobanu, V. G., Marcolli, C., Krieger, U. K., Zuend, A., and Peter, T.: Efflorescence of Ammonium Sulfate and Coated Ammonium Sulfate Particles: Evidence for Surface Nucleation, J. Phys. Chem. A, 114, 9486–9495, https://doi.org/10.1021/jp103541w, 2010.

Fredenslund, A., Jones, R. L., and Prausnitz, J. M.: Group-contribution estimation of activity coefficients in nonideal liquid mixtures, AIChE Journal, 21, 1086–1099, https://doi.org/10.1002/aic.690210607, 1975.

Gervasi, N. R., Topping, D. O., and Zuend, A.: A predictive group-contribution model for the viscosity of aqueous organic aerosol, Atmospheric Chemistry and Physics, 20, 2987–3008, https://doi.org/10.5194/acp-20-2987-2020, 2020.

Hansen, H. K., Rasmussen, P., Fredenslund, A., Schiller, M., and Gmehling, J.: Vapor-liquid equilibria by UNIFAC group contribution. 5. Revision and extension, Ind. Eng. Chem. Res., 30, 2352–2355, https://doi.org/10.1021/ie00058a017, 1991.

Hodas, N., Zuend, A., Schilling, K., Berkemeier, T., Shiraiwa, M., Flagan, R. C., and Seinfeld, J. H.: Discontinuities in hygroscopic growth below and above water saturation for laboratory surrogates of oligomers in organic atmospheric aerosols, Atmospheric Chemistry and Physics, 16, 12767–12792, https://doi.org/10.5194/acp-16-12767-2016, 2016.

Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, Phys. Chem. Chem. Phys., 13, 19238–19255, https://doi.org/10.1039/C1CP22617G, 2011.

Lilek, J. and Zuend, A.: A predictive viscosity model for aqueous electrolytes and mixed organic–inorganic aerosol phases, Atmospheric Chemistry and Physics, 22, 3203–3233, https://doi.org/10.5194/acp-22-3203-2022, 2022.

Marcolli, C. and Krieger, U. K.: Phase changes during hygroscopic cycles of mixed organic/inorganic model systems of tropospheric aerosols, J. Phys. Chem. A, 110, 1881–1893, https://doi.org/10.1021/jp0556759, 2006.

Shiraiwa, M., Ammann, M., Koop, T., and Pöschl, U.: Gas uptake and chemical aging of semisolid organic aerosol particles, Proc Natl Acad Sci U S A, 108, 11003–11008, https://doi.org/10.1073/pnas.1103045108, 2011.

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.

Wang, L.-N., Cai, C., and Zhang, Y.-H.: Kinetically Determined Hygroscopicity and Efflorescence of Sucrose-Ammonium Sulfate Aerosol Droplets under Lower Relative Humidity, J Phys Chem B, 121, 8551–8557, https://doi.org/10.1021/acs.jpcb.7b05551, 2017.

Zuend, A., Marcolli, C., Luo, B. P., and Peter, T.: A thermodynamic model of mixed organic-inorganic aerosols to predict activity coefficients, Atmospheric Chemistry and Physics, 8, 4559–4593, https://doi.org/10.5194/acp-8-4559-2008, 2008.

Zuend, A., Marcolli, C., Booth, A. M., Lienhard, D. M., Soonsin, V., Krieger, U. K., Topping, D. O., McFiggans, G., Peter, T., and Seinfeld, J. H.: New and extended parameterization of the thermodynamic model AIOMFAC: calculation of activity coefficients for organic-inorganic mixtures containing carboxyl, hydroxyl, carbonyl, ether, ester, alkenyl, alkyl, and aromatic functional groups, Atmospheric Chemistry and

Physics, 11, 9155–9206, https://doi.org/10.5194/acp-11-9155-2011, 2011.

Referee 5:

We would like to sincerely thank the editor and the reviewers for their thorough assessment of our manuscript and for their valuable comments and suggestions. We have carefully revised the manuscript in response to all comments, and we believe that these revisions have significantly improved the manuscript. In the following, we provide a detailed point-by-point response, with all changes incorporated into the revised version.

1. 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.

Response: We thank the reviewer for this helpful comment. Following the reviewer's suggestion, we have revised the manuscript accordingly. We moved the relevant discussion to the introduction. In addition, we also revised the opening part of section 3.1 and added the following clarification.

Revision: *(1) Introduction: The efflorescence process of atmospheric aerosols is primarily controlled by the nucleation and crystal growth (i.e., crystallization) of inorganic salts within aerosol particles, but some organics can also exist as crystalline solid, i.e., undergo crystallization/efflorescence. (2) Section 3.1: In the study of ERH presented here, we focus on the efflorescence of the inorganic component in internally mixed organic-inorganic aerosols.*

2. 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?

Response: We sincerely thank the reviewer for pointing this out. In the revised manuscript, we have added a more detailed description of the efflorescence process and clarified the complementary roles of thermodynamics and kinetics. We now state that thermodynamic supersaturation is a prerequisite for efflorescence, while the observed ERH is further modulated by kinetic barriers to nucleation and crystal growth, which are strongly influenced by aerosol viscosity. We have also clarified that the ERH model presented here is an empirical parameterization based on experimentally reported ERH values, and therefore represents the average/representative onset behavior of efflorescence rather than the full statistical distribution of crystallization events.

Revision: ***(1)*** *Thermodynamics-based approaches, such as that described by Hodas et al. (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.

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; Kuwata and Martin, 2012), 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 still 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 particle size, phase morphology, and viscosity.

***(2)** In addition, the ERH model presented here is an empirical parameterization based on experimentally reported ERH values; therefore, it is intended to capture the occurrence of efflorescence, rather than the full statistical distribution of crystallization events.*

More information on the ERH-viscosity model (eqs. 4 and 5) is required.

3. 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.

Response: We sincerely thank the reviewer for highlighting the need for these important methodological details. We fully agree that both temperature and particle size are critical variables that influence ERH and viscosity, and their specific ranges must be explicitly stated to properly define the model's boundary conditions. Regarding

temperature, we have added explicit statements emphasizing that our model is currently limited to standard room temperature conditions. Regarding particle size, the majority of the empirical ERH data, particularly within the training set, originates from our research group's previous laboratory observations. The particles investigated in these experimental studies primarily fall within the micrometer size range, typically between 1 and 10 μm. We have incorporated these detailed descriptions into the revised manuscript to clarify the experimental origins of our dataset and the applicable scope of the regression model.

Revision: ***Section 2.1:*** *The particles investigated in our previous studies typically range from 1 to 10 μm in diameter.*

Section 3.2: *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.*

4. 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).

Response: We sincerely thank the reviewer for raising this point. Our splitting procedure was a separation based on the source of the 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 research group, 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.

Detailed explanations have been added in the method part for further clarification.

5. 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 η, 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?

Response: We thank the reviewer for this valuable comment. In the revised manuscript, we have expanded the discussion of the model uncertainty, the assumptions underlying the linear regression, and the range of conditions over which the model may be applied. First, we clarified that the dark pink shading in Figures 2 and 3 represents the 95% confidence interval of the regression, whereas the light pink shading denotes the 95% prediction interval. The confidence interval reflects the uncertainty in the fitted mean relationship, while the prediction interval represents the expected spread of individual observations around the regression and is therefore wider.

In addition, we further clarify that this model is an empirical linear parameterization based on literature-reported ERH values and AIOMFAC-predicted viscosities, and is intended to represent the average or representative onset behavior of efflorescence rather than the full statistical distribution of crystallization events. Furthermore, we now explicitly state that the data used to develop this model were obtained at 298 ± 10 K; therefore, the model is only applicable to this near-room-temperature range, which is much narrower than the full range of temperatures encountered in the troposphere. Finally, we also clarify that the current model is mainly applicable to internally mixed organic–inorganic aerosols containing ammonium

sulfate, ammonium nitrate, sodium chloride, or potassium chloride, with organic components consisting primarily of water-soluble C, H, and O containing compounds, such as dicarboxylic acids and sugars.

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. In addition, the ERH model presented here is an empirical parameterization based on experimentally reported ERH values; therefore, it is intended to capture the occurrence of efflorescence, rather than the full statistical distribution of crystallization events. The model uncertainty reflects the scatter of the experimental ERH data, the stochastic nature of nucleation, and the uncertainty in AIOMFAC-predicted viscosity. The current model is mainly applicable to internally mixed organic–inorganic aerosols containing ammonium sulfate, ammonium nitrate, sodium chloride, or potassium chloride, with organic components primarily consisting of water-soluble C, H, and O, containing compounds, such as dicarboxylic acids and sugars.*

6. 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)".

Response: We thank the reviewer for pointing this out. We have carefully checked the manuscript and corrected the reference formatting throughout the text, including the missing spaces between the main text and citations.

7. 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.

8. 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)".

9. 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.

Response:7-9: We thank the reviewer for these careful comments. We have carefully checked the entire manuscript and corrected the punctuation inconsistencies, duplicate references, formatting problems in the bibliography, and the typos and notation inconsistencies throughout the text.

10. Section 3.3 "Nucleation kinetics": This section requires improvement and an overall more detailed discussion.

Response: We thank the reviewer for this comment. We have revised Section 3.3 accordingly and added a more detailed discussion.

Revision: Thermodynamics-based approaches, such as that described by Hodas et al. (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.

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; Kuwata and Martin, 2012), 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 still 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 particle size, phase morphology, and viscosity.

11. *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.*

Response: We thank the reviewer for this helpful comment. Most of the experiments included in this study were conducted in our laboratory using broadly consistent dehydration protocols, with the relative humidity decreasing at a rate of less than 0.1% s^{-1}, thereby ensuring good consistency among the measurements. Therefore, the dehumidification conditions were generally comparable for most of the data used to develop the model, although minor differences among studies may still contribute to part of the scatter in the dataset. Because most of the dataset was generated using this consistent protocol, the dehumidification rates were broadly comparable across the majority of the experiments used to build the model.

Following the reviewer's suggestion, we have added the available information on dehumidification conditions to the SI tables and clarified in the manuscript that the consistency of the dehumidification protocol across most of the dataset helps reduce uncertainty associated with kinetic effects.

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.***"**

12. L256: "W(T)/KT correlates with -logn". Please define and quantify "correlate". Also, you switch back and forth between log10 and In logarithm. Please introduce a consistency upon revision.

Response: We thank the reviewer for this careful comment. In the revised manuscript, we have made the corresponding changes to clarify this point and to use consistent logarithmic notation throughout the relevant discussion.

Revision:

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

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

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

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

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

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 $log_{10}v - \frac{\Delta G(T)}{2.303kT}$. *Overall, the viscosity-ERH model parameterization aligns with this framework, showing that aerosol ERH decreases with increasing* $log_{10}\eta$, *corresponding to reduced atomic rearrangement rates.*

13. 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 another 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.

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. Given that the broad implications of viscosity parameterization for global climate models remain subject to ongoing debate and have not yet been clearly established, we have removed this contentious statement from the revised manuscript.

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. ... 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."

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.

Response: We thank the reviewer for this careful comment. We have revised the text accordingly and now distinguish this type of hysteresis from the pronounced discontinuous hysteresis associated with efflorescence and deliquescence of crystalline particles.

Revision: *In contrast, amorphous organic aerosols generally do not exhibit the pronounced discontinuous hysteresis associated with efflorescence and deliquescence of crystalline particles. However, semi-solid or glassy amorphous particles may still show gradual, kinetically driven hysteresis in water uptake and release because of slow diffusion and equilibration. (Koop et al., 2011)*

L47: "introducing substantial uncertainties...". Please specify uncertainties, provide examples and appropriate references.

Response: We thank the reviewer for this comment. We have revised the text to specify that the uncertainties mainly concern the prediction of mixed aerosol water uptake and particle size under varying RH conditions. We also added references showing that phase transitions and kinetic limitations during hygroscopic cycles can lead to different water contents and growth factors at the same RH.

Revision: *Thus, effloresced (or crystallized) aerosols display fundamentally different hygroscopic behaviors from amorphous ones, introducing uncertainties in predicting mixing aerosol water uptake and particle size under varying RH conditions because phase transitions and kinetic limitations can lead to different water contents and growth factors during hygroscopic cycles. (Marcolli and Krieger, 2006; Mikhailov et al., 2009)*

L58-61: The statement should be supported by references.

Response: We thank the reviewer for this comment. Relevant references have been added in the revised manuscript.

L70: "is widely used". Since you argue that it is widely used, it would be appropriate to give more than one reference here.

Response: *We thank the reviewer for this comment. Relevant references have been added in the revised manuscript.*

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?

Response: We thank the reviewer for this helpful comment. In the revised manuscript, we have added more specific information on the AIOMFAC-VISC model and on how the viscosities were determined in this study. Regarding the uncertainty of the modelled viscosities, we now clarify that these values were not directly measured but predicted by AIOMFAC-VISC, and therefore carry uncertainty associated with thermodynamic state prediction and compositional variability. To assess this uncertainty, the model performed a sensitivity analysis in which the aerosol water mass fraction was perturbed by ±2%, and the resulting variation in predicted viscosity, expressed as $\pm\log_{10}(\eta/[\mathrm{Pa\cdot s}])$, was used as a proxy uncertainty metric. The corresponding values have been listed in Table S1.

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 (e.g., Figure 11 in Lilek and Zuend et al.) 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)

L156: Please provide a more detailed discussion for your choice of k_GT.

Response: We thank the reviewer for this insightful comment. The assumption of k_{GT} = 2.5 for all organic–water mixtures is an estimate based on established practices in the literature for secondary organic aerosol components. While the exact k_{GT} value depends on specific solute–water interactions, and can be precisely determined for well-investigated individual systems (e.g., k_{GT} =3.18 for citric acid), obtaining compound-specific parameters for every constituent within highly complex SOA mixtures presents practical challenges. In the absence of explicit experimental data for all constituent compounds, k_{GT} =2.5 is widely utilized as a robust and representative baseline derived from structurally related water-soluble organics. We have explicitly justified this assumption in the revised manuscript and acknowledged the associated uncertainty (typically evaluated as ±1.0) 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±1.0). This variability introduces a potential, albeit minor, source of uncertainty into the T_g-based model predictions presented in this study.*

Fig. 3: The x-axes are wrong/switched.

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".

References

Ciobanu, V. G., Marcolli, C., Krieger, U. K., Zuend, A., and Peter, T.: Efflorescence of Ammonium Sulfate and Coated Ammonium Sulfate Particles: Evidence for Surface Nucleation, J. Phys. Chem. A, 114, 9486–9495, https://doi.org/10.1021/jp103541w, 2010.

Fredenslund, A., Jones, R. L., and Prausnitz, J. M.: Group-contribution estimation of activity coefficients in nonideal liquid mixtures, AIChE Journal, 21, 1086–1099, https://doi.org/10.1002/aic.690210607, 1975.

Gervasi, N. R., Topping, D. O., and Zuend, A.: A predictive group-contribution model for the viscosity of aqueous organic aerosol, Atmospheric Chemistry and Physics, 20, 2987–3008, https://doi.org/10.5194/acp-20-2987-2020, 2020.

Hansen, H. K., Rasmussen, P., Fredenslund, A., Schiller, M., and Gmehling, J.: Vapor-liquid equilibria by UNIFAC group contribution. 5. Revision and extension, Ind. Eng. Chem. Res., 30, 2352–2355, https://doi.org/10.1021/ie00058a017, 1991.

Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, Phys. Chem. Chem. Phys., 13, 19238–19255, https://doi.org/10.1039/C1CP22617G, 2011.

Kuwata, M. and Martin, S. T.: Phase of atmospheric secondary organic material affects its reactivity, Proc Natl Acad Sci U S A, 109, 17354–17359, https://doi.org/10.1073/pnas.1209071109, 2012.

Lilek, J. and Zuend, A.: A predictive viscosity model for aqueous electrolytes and mixed organic–inorganic aerosol phases, Atmospheric Chemistry and Physics, 22, 3203–3233, https://doi.org/10.5194/acp-22-3203-2022, 2022.

Marcolli, C. and Krieger, U. K.: Phase changes during hygroscopic cycles of mixed organic/inorganic model systems of tropospheric aerosols, J. Phys. Chem. A, 110, 1881–1893, https://doi.org/10.1021/jp0556759, 2006.

Mikhailov, E., Vlasenko, S., Martin, S. T., Koop, T., and Pöschl, U.: Amorphous and crystalline aerosol particles interacting with water vapor: conceptual framework and experimental evidence for restructuring, phase transitions and kinetic limitations, Atmospheric Chemistry and Physics, 9, 9491–9522, https://doi.org/10.5194/acp-9-9491-2009, 2009.

Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nat Commun, 8, 15002, https://doi.org/10.1038/ncomms15002, 2017.

Zuend, A., Marcolli, C., Luo, B. P., and Peter, T.: A thermodynamic model of mixed organic-inorganic aerosols to predict activity coefficients, Atmospheric Chemistry and Physics, 8, 4559–4593, https://doi.org/10.5194/acp-8-4559-2008, 2008.

Zuend, A., Marcolli, C., Booth, A. M., Lienhard, D. M., Soonsin, V., Krieger, U. K., Topping, D. O., McFiggans, G., Peter, T., and Seinfeld, J. H.: New and extended parameterization of the thermodynamic model AIOMFAC: calculation of activity coefficients for organic-inorganic mixtures containing carboxyl, hydroxyl, carbonyl, ether, ester, alkenyl, alkyl, and aromatic functional groups, Atmospheric Chemistry and Physics, 11, 9155–9206, https://doi.org/10.5194/acp-11-9155-2011, 2011.

Referee 6

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 shirt 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 Tg and so the association between ERH and Tg 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):*

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

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:*

$$\frac{-W(T)}{kT} = -Alog_{10}\eta \qquad (10)$$

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

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

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.

Capouet, M. and Müller, J.-F.: A group contribution method for estimating the vapour pressures of α-pinene oxidation products, Atmospheric Chemistry and Physics, 6, 1455–1467, https://doi.org/10.5194/acp-6-1455-2006, 2006.

Cohen, M. D., Flagan, R. C., and Seinfeld, J. H.: Studies of concentrated electrolyte solutions using the electrodynamic balance. 3. Solute nucleation, J. Phys. Chem., 91, 4583–4590, https://doi.org/10.1021/j100301a031, 1987.

Davis, R. D., Lance, S., Gordon, J. A., Ushijima, S. B., and Tolbert, M. A.: Contact efflorescence as a pathway for crystallization of atmospherically relevant particles, Proc Natl Acad Sci U S A, 112, 15815–15820, https://doi.org/10.1073/pnas.1522860113, 2015.

Fitzgerald, C., Hosny, N. A., Tong, H., Seville, P. C., Gallimore, P. J., Davidson, N. M., Athanasiadis, A., Botchway, S. W., Ward, A. D., Kalberer, M., Kuimova, M. K., and Pope, F. D.: Fluorescence lifetime imaging of optically levitated aerosol: a technique to quantitatively map the viscosity of suspended aerosol particles, Phys Chem Chem Phys, 18, 21710–21719, https://doi.org/10.1039/c6cp03674k, 2016.

Galeazzo, T. and Shiraiwa, M.: Predicting glass transition temperature and melting point of organic compounds via machine learning and molecular embeddings, https://doi.org/10.1039/D1EA00090J, 2022.

Gupta, D., Kim, H., Park, G., Li, X., Eom, H.-J., and Ro, C.-U.: Hygroscopic properties of NaCl and $NaNO_3$ mixture particles as reacted inorganic sea-salt aerosol surrogates, Atmospheric Chemistry and Physics, 15, 3379–3393, https://doi.org/10.5194/acp-15-3379-2015, 2015.

Hollomon, J. H. and Turnbull, D.: Nucleation, Progress in Metal Physics, 4, 333–388, https://doi.org/10.1016/0502-8205(53)90020-3, 1953.

Hosny, N. A., Fitzgerald, C., Vyšniauskas, A., Athanasiadis, A., Berkemeier, T., Uygur, N., Pöschl, U., Shiraiwa, M., Kalberer, M., Pope, F. D., and Kuimova, M. K.: Direct imaging of changes in aerosol particle viscosity upon hydration and chemical aging, Chem. Sci., 7, 1357–1367, https://doi.org/10.1039/C5SC02959G, 2016.

Ji, Z.-R., Zhang, Y., Pang, S.-F., and Zhang, Y.-H.: Crystal Nucleation and Crystal Growth and Mass Transfer in Internally Mixed Sucrose/NaNO3 Particles, J. Phys. Chem. A, 121, 7968–7975, https://doi.org/10.1021/acs.jpca.7b08004, 2017.

Kelton, K. F., Greer, A. L., and Thompson, C. V.: Transient nucleation in condensed systems, J. Chem. Phys., 79, 6261–6276, https://doi.org/10.1063/1.445731, 1983.

Koop, T., Bookhold, J., Shiraiwa, M., and Pöschl, U.: Glass transition and phase state

of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere, Phys. Chem. Chem. Phys., 13, 19238–19255, https://doi.org/10.1039/C1CP22617G, 2011.

Li, Y., Pöschl, U., and Shiraiwa, M.: Molecular corridors and parameterizations of volatility in the chemical evolution of organic aerosols, Atmospheric Chemistry and Physics, 16, 3327–3344, https://doi.org/10.5194/acp-16-3327-2016, 2016.

Li, Y., Day, D. A., Stark, H., Jimenez, J. L., and Shiraiwa, M.: Predictions of the glass transition temperature and viscosity of organic aerosols from volatility distributions, Atmospheric Chemistry and Physics, 20, 8103–8122, https://doi.org/10.5194/acp-20-8103-2020, 2020.

Lilek, J. and Zuend, A.: A predictive viscosity model for aqueous electrolytes and mixed organic–inorganic aerosol phases, Atmospheric Chemistry and Physics, 22, 3203–3233, https://doi.org/10.5194/acp-22-3203-2022, 2022.

Ma, S., Pang, S., Li, J., and Zhang, Y.: A review of efflorescence kinetics studies on atmospherically relevant particles, Chemosphere, 277, 130320, https://doi.org/10.1016/j.chemosphere.2021.130320, 2021.

Mikhailov, E., Vlasenko, S., Martin, S. T., Koop, T., and Pöschl, U.: Amorphous and crystalline aerosol particles interacting with water vapor: conceptual framework and experimental evidence for restructuring, phase transitions and kinetic limitations, Atmospheric Chemistry and Physics, 9, 9491–9522, https://doi.org/10.5194/acp-9-9491-2009, 2009.

Pankow, J. F. and Asher, W. E.: SIMPOL.1: a simple group contribution method for predicting vapor pressures and enthalpies of vaporization of multifunctional organic compounds, Atmospheric Chemistry and Physics, 8, 2773–2796, https://doi.org/10.5194/acp-8-2773-2008, 2008.

Sheldon, C. S., Salazar, J., Palacios Diaz, T., Morton, K., Davis, R. D., and Davies, J. F.: Correlating the Viscosity and Rate of Water Diffusion in Semisolid Gel-Forming Aerosol Particles, ACS Earth Space Chem., 7, 2311–2320, https://doi.org/10.1021/acsearthspacechem.3c00241, 2023.

Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nat Commun, 8, 15002, https://doi.org/10.1038/ncomms15002, 2017.

Turnbull, D.: Under what conditions can a glass be formed?, Contemporary Physics, https://doi.org/10.1080/00107516908204405, 1969.

Wang, L.-N., Cai, C., and Zhang, Y.-H.: Kinetically Determined Hygroscopicity and Efflorescence of Sucrose-Ammonium Sulfate Aerosol Droplets under Lower Relative

Humidity, J Phys Chem B, 121, 8551–8557, https://doi.org/10.1021/acs.jpcb.7b05551, 2017.

Zobrist, B., Marcolli, C., Pedernera, D. A., and Koop, T.: Do atmospheric aerosols form glasses?, Atmospheric Chemistry and Physics, 8, 5221–5244, https://doi.org/10.5194/acp-8-5221-2008, 2008.