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.

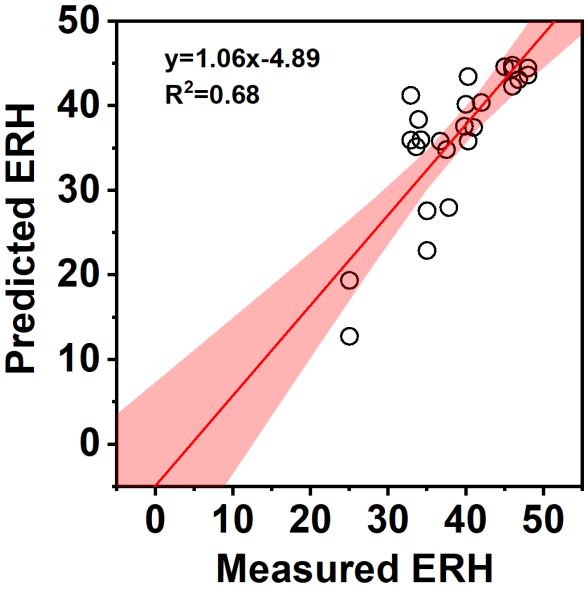
Table R1 *has been added to the Supplementary Information.*

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

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