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

Dear. Dr. Giorio,

On behalf of my co-authors, I'm pleased to submit a revised version of our manuscript, entitled "Predicting Organic–Inorganic Aerosol Efflorescence Using Thermodynamically Modeled Viscosity", for further consideration in *Atmospheric Chemistry and Physics*. We sincerely appreciate the time and effort invested by you and the reviewers in evaluating our manuscript. We have carefully considered all comments and revised the manuscript accordingly. A point-by-point response is provided in the accompanying response letter, with all changes highlighted in the marked manuscript.

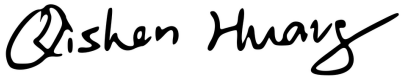
A brief summary of our responses is provided below:

For Reviewer Report 2 (Referee #4), we adopted the suggested use of the O:C ratio of the organic component and incorporated additional clarifications and references to improve accuracy and readability.

The major focus of this revision was to Reviewer Report 3 (Referee #2), which raised concerns regarding the interpretation of the viscosity–ERH relationship and the use of viscosity as a standalone predictor of aerosol ERH. In response, we now include the complete results of the additional analyses previously performed, including fixed-RH viscosity analyses. These results show that the viscosity–ERH correlation is weakened when RH is held constant, indicating that the RH dependence of aerosol viscosity contributes to the observed relationship. At the same time, modest viscosity-ERH correlations remain at intermediate RH values relevant to typical aerosol ERH, suggesting that a contribution of viscosity to efflorescence behavior cannot be excluded and warrants future investigation. Accordingly, we agree that viscosity should not be interpreted as a standalone predictor of ERH and have revised the manuscript throughout to reflect this point. We now present the viscosity–ERH relationship as an empirical viscosity–humidity framework that characterizes efflorescence behavior and defines phase-transition boundaries in viscosity–humidity space. The framework identifies distinct aqueous, efflorescence, and non-efflorescence regions, thereby providing quantitative constraints on aerosol phase transitions and hygroscopic hysteresis across a broad range of internally mixed organic–inorganic aerosol systems.

We believe these revisions have improved both the scientific rigor and clarity of the manuscript. We hope that the revised manuscript and detailed responses adequately address the reviewers' concerns. We sincerely appreciate your consideration and thank you again for the opportunity to revise our manuscript.

Sincerely,



Qishen Huang

Referee Report 1 (Referee #1):

For final publication, the manuscript should be accepted as is.

Response: We sincerely thank the reviewer for the positive evaluation of our manuscript and for supporting its publication in Atmospheric Chemistry and Physics. We greatly appreciate the reviewer's constructive comments and valuable suggestions, which have helped us improve the clarity, rigor, and overall quality of the manuscript. We are grateful for the time and effort the reviewer has devoted to assessing our work.

Referee Report 2 (Referee #4):

The authors received comments and suggestions from multiple reviewers and community comments and have responded and revised their manuscript accordingly. My own comments from the first round of reviews have been addressed adequately. Overall, the revised manuscript is a major improvement.
I have a few follow-up comments on some of the revised text. The line numbers referred to are those from the revised manuscript version.

Response: We sincerely thank the reviewer for the positive evaluation of our revised manuscript and for recognizing the substantial improvement made during the revision process. We are also grateful for the reviewer's careful follow-up comments on the revised text. We have carefully considered each of these suggestions and further revised the corresponding parts of the manuscript accordingly. We believe that these additional revisions have improved the clarity, accuracy, and overall quality of the manuscript.

1) Line 217: Phrasing of "However, the glass transition temperature of inorganic compounds is limited, …." should be improved. What the authors' likely mean is that the experimental data of glass transition temperatures of inorganic compounds are scarce (or limited) [the temperature itself is not limited].

Response: We thank the reviewer for pointing this out. We have revised the sentence accordingly to improve clarity and accuracy.

Revision: Line217: *"However, the experimental data of 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."*

2) Line 141 and Eq. (1): The given definition for "equivalent O:C ratio" is unfamiliar to this reviewer and seems odd. It would benefit from providing more context and reasoning for why this definition is meaningful (if it is).

While it is straightforward to define the average O:C ratio of a mixture of organic compounds (which Eq. (1) doesn't do), it seems odd to define an equivalent O:C ratio that is impacted by the inorganic solute amounts through x_org. Why not simply compute the O:C ratio of the organics? Is there a prior reference for defining "equivalent O:C ratio" this way or is this a new definition introduced in this study?

Eq. (1) implies that the presence of a high amount of inorganic solute leads to a low equivalent O:C ratio. In the context of organic mixtures and water uptake (and thus influence on viscosity), a low O:C ratio would indicate poor miscibility with water and poor hygroscopicity, but a low equivalent O:C ratio due to low x_org may mean the opposite. A low equivalent O:C ratio due to low x_org could apply to a particle as a whole that is quite hygroscopic (given the large n_inorg) and the actual organics present may even have a high O:C ratio. In contrast, the equivalent O:C ratio could also be low because the O:C ratios of the organics involved are low while x_org is high. The resulting physicochemical properties in that case would differ substantially from a case with high-O:C organics but low x_org. Therefore, the physicochemical meaning and usefulness of the given definition is questionable.

Response: We thank the reviewer for raising this important question. The systems involved in this study are all ternary systems with one organic, one inorganic salt, and water, thus average O:C is not applicable in this study. We therefore intended to define the equivalent O:C to account for the change in organic fractions; however, the contrasting direction for increasing organic fraction and increasing polarity intended by the equivalent O:C was neglected. Thus, to avoid ambiguity and to provide a clearer

physicochemical meaning, we have revised the manuscript to directly use the O:C ratio of the organic compound itself rather than the previously defined equivalent O:C ratio. The results of multivariate model was revised accordingly. The multivariate model also accounts for the contribution of organic fractions, but with the change from equivalent O:C to O:C the major findings in comparing the viscosity-ERH model and the multivariate model were not affected. Corresponding revisions have been made in the main text at Line 150, Figure 1d, Table S1, and Table S8 – S11.

Revision: Line 16: *"Herein, we evaluated several chemical parameters, including O:C ratio, organic mass fractions, glass transition temperature (T_g), and viscosity (η), … "*

Line 146: "*Key physicochemical parameters for describing aerosol ERH include the O:C ratio of the organic compound in the mixture:*

$$O{:}C = \frac{N_{O,org}}{N_{C,org}} \quad (1)$$

where $N_{O,org}$ and $N_{C,org}$ are the number of oxygen and carbon atoms in the organic compound, respectively. Other parameters include ω_{org}, fractional T_g at ERH ($T_{g(\omega org)}$), and viscosity."

Figure 1d:

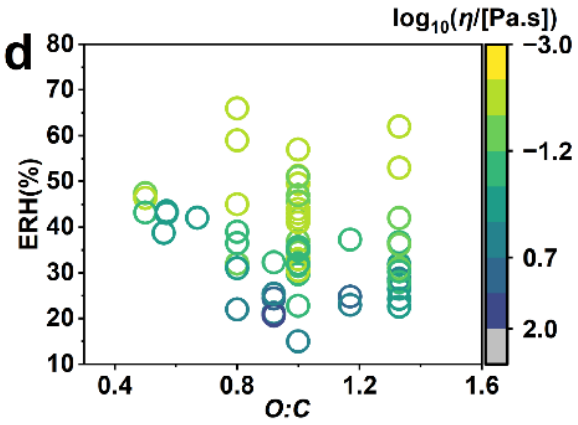


Figure 1. Relationships between the ERH of organic-inorganic mixed aerosols. (a) the logarithm of viscosity (log10(η/[Pa.s])), (b) the organic mass fraction (ωorg), (c) the glass transition temperature (Tg(ωorg), K) calculated using the methods of Li et al. (Li et al., 2020) and Armeli et al. (Armeli et al., 2023), respectively, and (d) O:C ratio. The color bar denotes the aerosol viscosity in (log10(η/[Pa.s])).

Other changes are made in Table S1, and Table S8 – S11.

3) Eq. (2): Please define the variables listed (n_inorg, n_org) and make clear that n_inorg does not included water. This was clarified in the authors' response document but not the revised manuscript.

Response: We thank the reviewer for this helpful comment. As described in our response to Comment 2, we have removed the previous definition of the "equivalent O:C ratio" and the corresponding equation from the revised manuscript. Therefore, the variables (n_{inorg}) and (n_{org}) are no longer used or defined in this section. The related revisions have been made accordingly, as detailed in our response to Comment 2.

4) Line 411: "supersaturation relative to the ion activity product (IAP)" is lacking important connection to a reference condition. What the authors mean is supersaturation relative to the ion activity product (IAP) at the point of saturation of the species involved in the crystalline phase (i.e., IAP_sat).

Response: We thank the reviewer for this helpful clarification. We agree that the previous wording was incomplete because the ion activity product (IAP) should be referenced to the saturation condition of the species involved in the crystalline phase. We have revised the relevant sentence at Line 428 to explicitly define supersaturation relative to the ion activity product at saturation, i.e., IAP_{sat}.

Revision: Line 433: "*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) at the point of saturation of the species involved in the crystalline phase (i.e., IAP_{sat}). 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.*"

5) Line 135: "typical atmospheric aerosol lifetimes (days to weeks)," Please add a reference like Anon et al. (2000) here; it was mentioned in the authors' response but not the revised manuscript.

Response: We thank the reviewer for this helpful suggestion. We agree that a reference should be added to support the statement regarding typical atmospheric aerosol lifetimes. We have now added the relevant citation in the revised manuscript at Line 142. We also note that the author name mentioned in our previous response was incorrect, and we have corrected it in the revised manuscript.

Revision: Line 139-141: "*Although these timescales are shorter than typical atmospheric aerosol lifetimes (days to weeks), previous studies have shown that nucleation induction times in high-viscosity systems can be extremely long due to diffusion limitations. (Raes, 2000; Wang et al., 2017)"*

6) Line 168: Likely an oversight: "Validation against …" is a redundant statement with that given in the preceding sentence.

Response: We thank the reviewer for pointing out this oversight. We agree that the previous text contained a redundant statement regarding the validation of the AIOMFAC-VISC approach. We have deleted the repeated sentence and retained a more concise description in the revised manuscript.

Revision: Line 167-172: *"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; Song et al., 2021; Zuend et al., 2011)"*

Referee Report 3 (Referee #2):

While I appreciate that the authors have substantively responded to my review and those of the other reviewers, it is my scientific opinion that the revisions have not fully grappled with the implications of the additional analyses that they performed in response to my prior concern. My prior review stated: "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."

To the authors' credit, they performed several analyses that I suggested. Regarding the first analysis regarding how much viscosity changes with RH for a given chemical composition, they acknowledged: "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."

For the second analysis looking into a correlation between the aerosol viscosity at a consistent RH (to be able to make a likewise comparison) and ERH, the author's acknowledged "In all cases, the correlation was significantly reduced when RH was held constant." Unfortunately, the authors declined to include any of this analysis in the main text or in the SI (as far as I could find), raising the question of what exactly the magnitude of the "significant reduction" might have been. Another statement in the response suggests that the relationship may have indeed been entirely eliminated, as they also stated elsewhere in their review that: "An intrinsic, independent correlation between modeled viscosity and ERH is not explicitly observed."

Overall, these analyses do not allay my concern that this work has not identified any mechanistic connection between viscosity and ERH, and has simply identified a correlation between two variables that are already well known to be strongly correlated (viscosity and RH). After performing all these analyses, the authors added this sentence to the main text:

"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."

I completely agree with the first part of the sentence. The second part ("and confirm that efflorescence is governed by nucleation and crystal growth kinetics controlled by viscosity at the ERH") I do not believe is at all justified by the presented data or analysis.

In my opinion, the simplest explanation of their data, that they are just looking at a correlation between viscosity and RH in a somewhat roundabout way, has not been disproven by these further analyses and in fact has been reinforced. If "efflorescence is governed by nucleation and crystal growth kinetics controlled by viscosity at the ERH", then wouldn't one expect viscosity at the ERH to be constant, or nearly constant? Instead, it varies over ~4 orders of magnitude.

While the authors dutifully performed these analyses and added a sentence or two of caveats to the main text, to me, the overall balance of the paper still heavily implies that there is a mechanistic connection between ERH and viscosity, and not simply a correlation. For instance, the final sentence in the abstract states: "Our findings highlight the role of aerosol viscosity in controlling efflorescence and emphasize the need to develop improved aerosol viscosity measurement techniques to better constrain aerosol phase transitions in atmospheric models."

At the risk of repeating myself too much, I still do not be a mechanistic connection (or "control" in the language of that sentence) has been established, simply a correlation.

Regarding the second half of this sentence, and looking to the bigger picture, I have one more point regarding the potential practical significance of this work. One might argue that a correlation can still be useful in a practical sense, which is certainly true in general. However, what has been correlated here is viscosity at the ERH, and ERH. Therefore, I see no way in which this relationship could be used to predict ERH, as that seems entirely circular (ERH could be predicted by the viscosity at the ERH, but to know viscosity at the ERH, obviously the ERH would have to known in the first place). The authors have already acknowledged that a relationship between viscosity more generally (not at each ERH) and ERH did not exist (or at least, the relationship is

"significantly reduced" and the exact relationship was not included).

Overall, then, in the absence of some further evidence or analysis that shows a deeper relationship between viscosity and ERH, it appears to me that the heart of this paper is simply the already well known correlation between viscosity and RH, which is already well established. Therefore, I do not recommend this paper for publication.

Response: We sincerely thank the reviewer for carefully re-evaluating our revised manuscript. The reviewer's suggestions helped us further refine our interpretation of the viscosity–ERH relationship. In this response, we take the opportunity to clarify several aspects that were not sufficiently presented or discussed in the previous revision.

During our previous revision, we performed both analyses suggested by the reviewer. However, while the results of analysis (i) were discussed in detail, the results of analysis (ii) were only briefly summarized in the manuscript. In response to the present comment, we have now added the complete results of both analyses to the Supplementary Information (Figures S7 and S8, shown in the Revision below), together with corresponding discussion in both the main text and SI.

More importantly, in response to the reviewer's concern, we have substantially revised the presentation and interpretation of the framework throughout the manuscript, including the abstract, discussions, conclusions, and implications. Specifically, the revised manuscript now: (1) explicitly presents and discusses the fixed-RH analysis; (2) reframes the viscosity–ERH relationship as an empirical viscosity–humidity framework defining phase-transition boundaries rather than evidence of a direct mechanistic relationship; and (3) clarifies the practical application of the framework as a phase-transition classification tool. Details are provided below.

(1) Results of the viscosity-humidity analyses

As noted in our previous revision, viscosity for a given chemical system varies strongly with RH. Therefore, the fitted relation between ERH and viscosity presented in the manuscript can be interpreted as an apparent representation of the combined effects of compositional variability across different aerosol compositions (Table S12 and Figure S7).

For the second analysis examining the relation between ERH and viscosity at fixed

RH, we evaluated viscosities at RH values from 20% to 90% and have now added the corresponding results to Figure S8 (shown below as Figure R1). Overall, as we described in our previous revision, the correlation between viscosity and ERH is greatly reduced when RH is held constant.

However, we found that the reduction is not uniform across RH. The highest correlations occur at intermediate RH values that overlap with the ERH range of many atmospheric aerosol systems (R^2 = 0.29 at RH = 60% and R^2 = 0.24 at RH = 50%), whereas substantially weaker correlations are observed at RH values well below or above the typical ERH range (e.g., R^2 = 0.15 at RH = 20% and R^2 = 0.08 at RH = 90%).

Although these correlations remain weak overall and are insufficient to establish an independent predictive relationship, they suggest that viscosity may still contribute to variations in observed ERH under conditions relevant to efflorescence. We therefore interpret these results as indicating that the RH dependence of viscosity explain the majority of the observed viscosity–ERH relationship, while the potential contribution from viscosity itself cannot be excluded and warrants further investigation.

(2) Revised interpretation of the model throughout the manuscript:

In light of these analyses, we agree that viscosity alone should not be viewed as an independent predictor of ERH. Accordingly, we have revised the manuscript throughout to avoid implying a direct mechanistic relationship between viscosity and ERH.

The viscosity–ERH framework is now described as an empirical representation of aerosol phase-transition behavior in viscosity–humidity space. Rather than relying on viscosity evaluated specifically at ERH, the framework can be applied using independently determined ambient RH and aerosol viscosity. The fitted relationship is interpreted as defining boundaries in viscosity–humidity space that separate three empirical regions: **Region I:** Above the fitted boundary line, where aerosols remain in an aqueous state. **Region II:** Below the boundary but with $\eta < 4.76\times10^2$ Pa·s, where nucleation and crystal growth leading to efflorescence are favored. **Region III:** $\eta > 4.76\times10^2$ Pa·s, where aerosols remain highly viscous and do not exhibit measurable efflorescence.

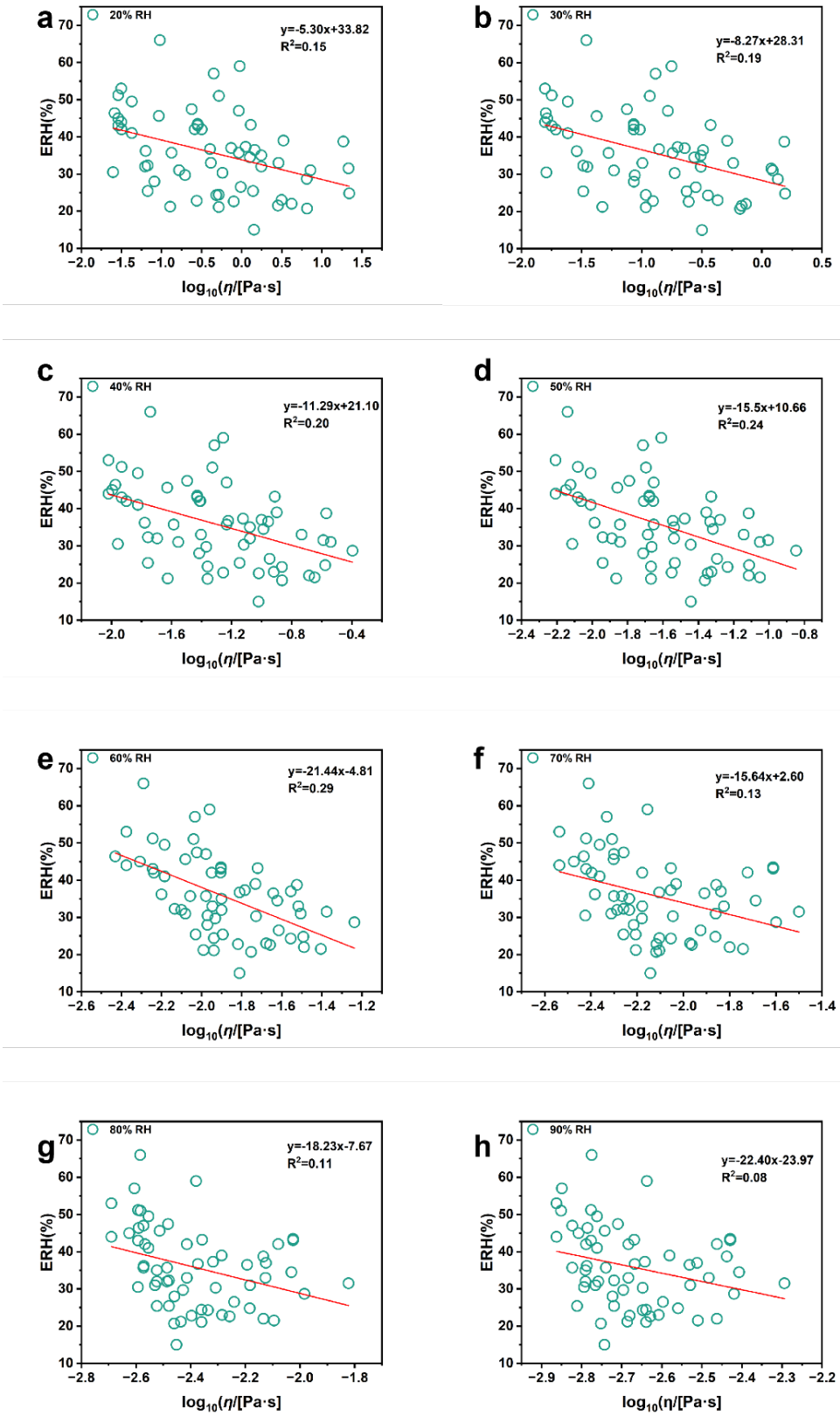


Figure R1. (Figure S8) *Relationships between ERH and aerosol viscosity calculated at fixed RH conditions. Panels (a–h) show ERH plotted against (log10(η/[Pa·s])), where η represents the aerosol viscosity calculated at fixed RH values of 20%, 30%, 40%, 50%, 60%, 70%, 80%, and 90%, respectively.*

(3) Revised practical significance of the framework

We agree with the reviewer that a direct correlation between viscosity at ERH and ERH would have limited practical utility because of its circular nature. This issue was carefully considered in our previous revision. However, we recognize that our presentation did not sufficiently communicate that the intended use of the framework was no longer as a standalone predictor of ERH, but rather as a means of interpreting aerosol phase-transition behavior within viscosity–humidity space.

The revised manuscript therefore no longer presents the fitted relationship as a standalone predictor of ERH. Instead, we interpret the viscosity–ERH space as defining empirical transition regions associated with different aerosol phase outcomes. In this framework, ambient RH and aerosol viscosity are treated as independent inputs, and their position relative to the empirical boundary is used to characterize the expected aerosol behavior (aqueous, efflorescing, or highly viscous/non-efflorescing).

We believe this revised interpretation more accurately reflects the strengths and limitations of the framework. Its primary value is not as a one-to-one predictor of ERH, but as a means of organizing and characterizing observed aerosol phase-transition behavior across diverse organic–inorganic systems. Whether viscosity can ultimately serve as a more direct mechanistic predictor of ERH remains an important topic for future investigation. The present analyses suggest a possible contribution of viscosity, particularly at RH values relevant to efflorescence, but the available evidence is not sufficiently conclusive to establish such a relationship.

Again, we sincerely thank the reviewer for raising this important conceptual issue. We believe that the additional analyses, together with the substantial revisions to the interpretation and presentation of the framework, have resulted in a more balanced and scientifically rigorous manuscript that more clearly communicates both the utility and limitations of the proposed viscosity–ERH framework.

Revision：

Line 15: *“quantitative descriptions of efflorescence behavior in organic–inorganic aerosols remain underdeveloped.”*

Line 18-27: *"We developed a linear viscosity ($log_{10}\eta$)–ERH framework, which defines boundary conditions for aerosol efflorescence regimes. Above the fitted line, aerosols remain in an aqueous state, whereas below the line, efflorescence occurs. Additionally, efflorescence is unlikely to occur when η exceeds a threshold value ($> 4.76 \times 10^2$ Pa·s). The predicted boundary line was validated using an independent dataset, and a multivariate regression model incorporating η and T_g improved the statistical performance of the boundary line but was limited by T_g parameterization for complex organic-inorganic mixtures. Our findings highlight the association between aerosol viscosity and efflorescence and emphasize the need to develop improved aerosol viscosity measurement techniques to better constrain aerosol phase behavior in atmospheric models."*

Line 72-77: *"While laboratory studies have provided substantial data on efflorescence in mixed organic-inorganic aerosols, a comprehensive understanding of the factors influencing efflorescence remains limited. (Cai et al., 2017; Ma et al., 2021b; Wang et al., 2017) Efflorescence involves the formation of crystalline nuclei from a supersaturated solution and proceeds through kinetically controlled nucleation and crystal growth that requires overcoming an energy barrier. This process is governed by a combination of thermodynamic driving forces and kinetic limitations."*

Line 87-88: *"Thermodynamic constraints further require that efflorescence and crystallization can only occur when the aqueous phase becomes supersaturated with respect to the relevant solid phase"*

Line 108-115: *"In this study, we develop a framework to predict efflorescence in organic–inorganic aerosols by integrating literature-reported ERH data and evaluating key physicochemical parameters, with particular emphasis on aerosol viscosity (η) and the combined effects of T_g, O:C ratio, and ω_{org}. This framework constrains the efflorescence phase transition in viscosity–humidity space and provides a quantitative description of boundaries separating aqueous, efflorescence and non-efflorescence states. The result improves the representation of aerosol phase behavior by using ambient RH and thermodynamically modeled viscosity, and offers constraints for atmospheric processes such as gas–particle partitioning, heterogeneous chemistry,*

and cloud formation."

Line 265-267: *"We also note that the RH dependence of aerosol viscosity can contribute in the observed viscosity-ERH correlation, and we will discuss this in detail in the following section."*

Line 273-278: *"**3.2 Characterizing Efflorescence Regimes in Viscosity-Humidity Space.** Building on the correlation between viscosity and mean ERH values presented in Figure 1, we develop a viscosity–ERH framework (Figure 2) that establishes quantitative boundary lines in viscosity–humidity space. The boundary lines distinguish efflorescence and non-efflorescence regions, providing a simplified yet physically constrained representation of aerosol phase-transition behavior. The fitted equation for the boundary line is …"*

Line 281-282: *"This indicates that aerosol viscosity serves as a useful descriptor of efflorescence behavior across diverse organic–inorganic systems."*

Line 287-292: *"Since the AIOMFAC calculation accounts for inorganic salts, these results suggest that the observed viscosity–ERH relationship is broadly consistent across the inorganic salts represented in this study.*

The resulting viscosity–ERH framework provides a quantitative boundary for characterizing efflorescence regimes in viscosity–humidity space and for assessing the likelihood of efflorescence under a given combination of viscosity and ambient RH."

Line 294-297: *"Within the dataset examined here, aerosols with viscosities above this threshold did not exhibit measurable efflorescence. Accordingly, this threshold can be interpreted as a practical boundary separating efflorescence and non-efflorescence regions, suggesting that organic-inorganic aerosols with viscosities exceeding 4.76×10^{2} Pa·s are unlikely to undergo efflorescence."*

Line 310-332: *"To further examine the structure underlying the observed relationship, we analyzed (i) viscosity variations for fixed chemical systems across RH (20–60%) and (ii) the relationship between viscosity evaluated at fixed RH (from 20% - 90%) and ERH across different compositions. The slopes of the viscosity–RH relationships for representative systems range from -10 to -60 (Table S12, Figure S7); therefore, the fitted relation between ERH and viscosity can be interpreted as an apparent representation*

of the combined effects of compositional variability across different aerosol compositions.

The correlation between viscosity and ERH is greatly reduced when RH is held constant (Figure S8). However, we found that the reduction is not uniform across RH. The highest correlations occur at intermediate RH values that overlap with the ERH range of many atmospheric aerosol systems (R^2 = 0.29 at RH = 60% and R^2 = 0.24 at RH = 50%), whereas substantially weaker correlations are observed at RH values well below or above the typical ERH range (e.g., R^2 = 0.15 at RH = 20% and R^2 = 0.08 at RH = 90%). Although these correlations remain weak overall and are insufficient to establish an independent predictive relationship, they suggest that viscosity may still contribute to variations in observed ERH under conditions relevant to efflorescence. We therefore interpret these results as indicating that the RH dependence of viscosity explain the majority of the observed viscosity–ERH relationship, while the potential contribution from viscosity itself cannot be excluded and warrants further investigation.

Accordingly, this consolidates the interpretation of the viscosity–ERH framework as an empirical boundary representation in viscosity–humidity space that captures variability arising from organic composition and organic–inorganic interactions. In this framework, aerosol viscosity is not regarded as a standalone predictor of ERH, rather, ambient RH and aerosol viscosity are treated as independent inputs, and their position relative to the fitted boundary is used to characterize the expected aerosol state (aqueous, efflorescing, or highly viscous/non-efflorescing).”

Line 343-346: *“In contrast, the viscosity–ERH framework achieves comparable explanatory capability while requiring fewer assumptions. As a result, it provides a simpler and more broadly applicable approach for characterizing efflorescence regimes and phase-transition boundaries across diverse internally mixed organic–inorganic aerosols.”*

Line 375-386: *“The viscosity-ERH relationship defines a boundary line and a threshold value in viscosity–humidity space, which can be used in combination with ambient RH value and aerosol viscosity to interpret aerosol phase behavior within three distinct regions (Figure 3a). In Region I (above the red regression line), aerosols remain*

in an aqueous state. In Region II (below the regression boundary but with $\eta < 4.76\times10^2$ *Pa·s), aerosols are kinetically favorable for nucleation and crystal growth, corresponding to efflorescence. In Region III (*$\eta > 4.76\times10^2$ *Pa·s), aerosols remain in a highly viscous state and do not exhibit measurable efflorescence, instead forming amorphous solids.*

To validate the viscosity–ERH boundary framework derived from linear regression (Eq. 6), we compared the experimentally measured ERH values with the boundary line RH estimated from the fitted relationship (Figure 3b). The data points cluster around the y = x reference line (black dashed line), with the fitted line (red solid line) showing close alignment and a slope near unity ($R^2 = 0.95$*), indicating strong consistency between the empirical ERH values and the viscosity–ERH boundary representation."*

Line 420-428: ***"3.3 Physical interpretation of the viscosity-ERH framework.***

...

The viscosity-ERH framework established in this study is qualitatively consistent with Eq. 10, in which increasing viscosity is associated with reduced molecular mobility and lower effective nucleation rates. In this context, $log_{10}\eta$ *can be viewed as a practical indicator for the kinetic energy barrier W(T) on nucleation and crystal growth. The intercept achieved in Eq.(5) can therefore be associated with* $log_{10}v - \frac{\Delta G(T)}{2.303kT}$. *However, as discussed in Section 3.2, the observed viscosity–ERH relationship is strongly influenced by the RH dependence of viscosity. Therefore, the present results should not be simply interpreted as establishing a unique mechanistic relationship between viscosity and ERH. Rather, the viscosity–ERH framework provides an empirical description of the conditions under which efflorescence may occur for a broad range of internally mixed organic–inorganic aerosol systems."*

Line 439-448: *"Under these conditions, efflorescence can be viewed as a coupled thermodynamic–kinetic process. Thermodynamic supersaturation provides the driving force necessary for crystallization, whereas nucleation and crystal growth remain subject to kinetic limitations. Thus, thermodynamic and kinetic perspectives are complementary: thermodynamic conditions determine whether crystallization is*

possible, while kinetic factors influence when crystallization occurs. In this context, aerosol viscosity is considered as a practical descriptor of molecular mobility and diffusion limitations, rather than a standalone determinant of efflorescence.

While the viscosity–ERH framework characterizes distinct efflorescence regimes across diverse aerosol systems, its application to predicting aerosol phase behavior in viscosity–humidity space has inherent limitations"

Line 453-457: *"Consequently, the occurrence of efflorescence is influenced by a combination of thermodynamic driving force, particle morphology, interfacial properties, particle size, and molecular mobility. The viscosity–ERH framework presented here should therefore be viewed as an empirical boundary description of observed aerosol behavior rather than a complete mechanistic model of crystallization."*

Line 459-468: *"Our results demonstrate that aerosol viscosity provides a useful framework for characterizing efflorescence behavior in internally mixed organic–inorganic aerosols. Although inclusion of both viscosity and T_g slightly improves model performance, the viscosity–ERH framework offers broader applicability and greater simplicity for diverse aerosol systems. The framework remains valuable for describing aerosol phase transitions because it organizes observed behavior into distinct regimes in viscosity–humidity space and defines a boundary separating different phase states. Within the dataset examined here, three behavioral regions are identified: (i) an aqueous regime above the fitted boundary line, (ii) an efflorescence regime below the boundary line when viscosity remains below the threshold value, and (iii) a non-efflorescence regime at viscosities exceeding approximately 4.76×10^2 Pa·s. This provides a physically constrained representation of aerosol phase-transition behavior."*

Line 496-499: *"Analyses in this study suggest a possible contribution of viscosity, particularly at RH values relevant to efflorescence, but the available evidence is not sufficiently conclusive to establish such a relationship. Therefore, whether viscosity can ultimately serve as a more direct mechanistic predictor of ERH remains an important topic for future investigation."*

Figure 3:

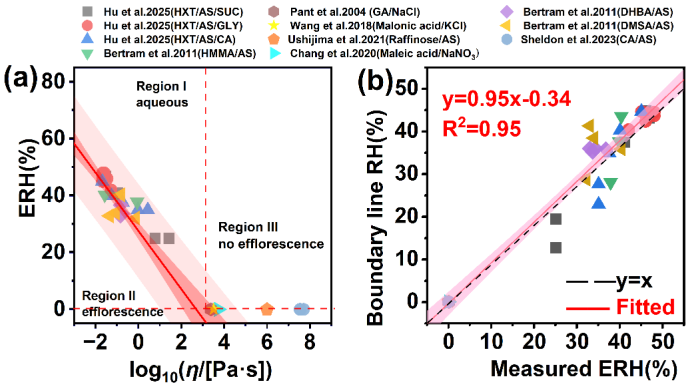


Figure 3. (a) Comparison between the validation dataset and viscosity-ERH boundary framework (Eq. 5), showing three distinct regions in viscosity–humidity space: Region I (aqueous state), Region II (efflorescence-favorable conditions), and Region III (non-efflorescence or highly viscous state). (b) Comparison between experimentally measured ERH values and the boundary line RH values derived from the viscosity–humidity relationship. The black dashed line denotes the y = x reference line, and the red line represents the linear regression fit to the data, along with the corresponding coefficient of determination, R2=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.

Figure S7:

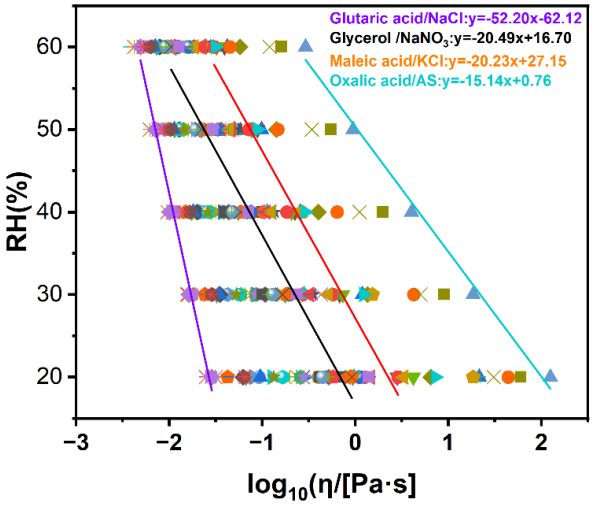


Figure S7. Relationship between relative humidity (RH) and aerosol viscosity for organic–inorganic mixed aerosol systems. The data points represent the modeled viscosities of different organic–inorganic aerosol compositions at selected RH values from 20% to 60%. The solid lines show linear fits for representative chemical systems, with the corresponding fitting equations shown in the figure.

Figure S8:

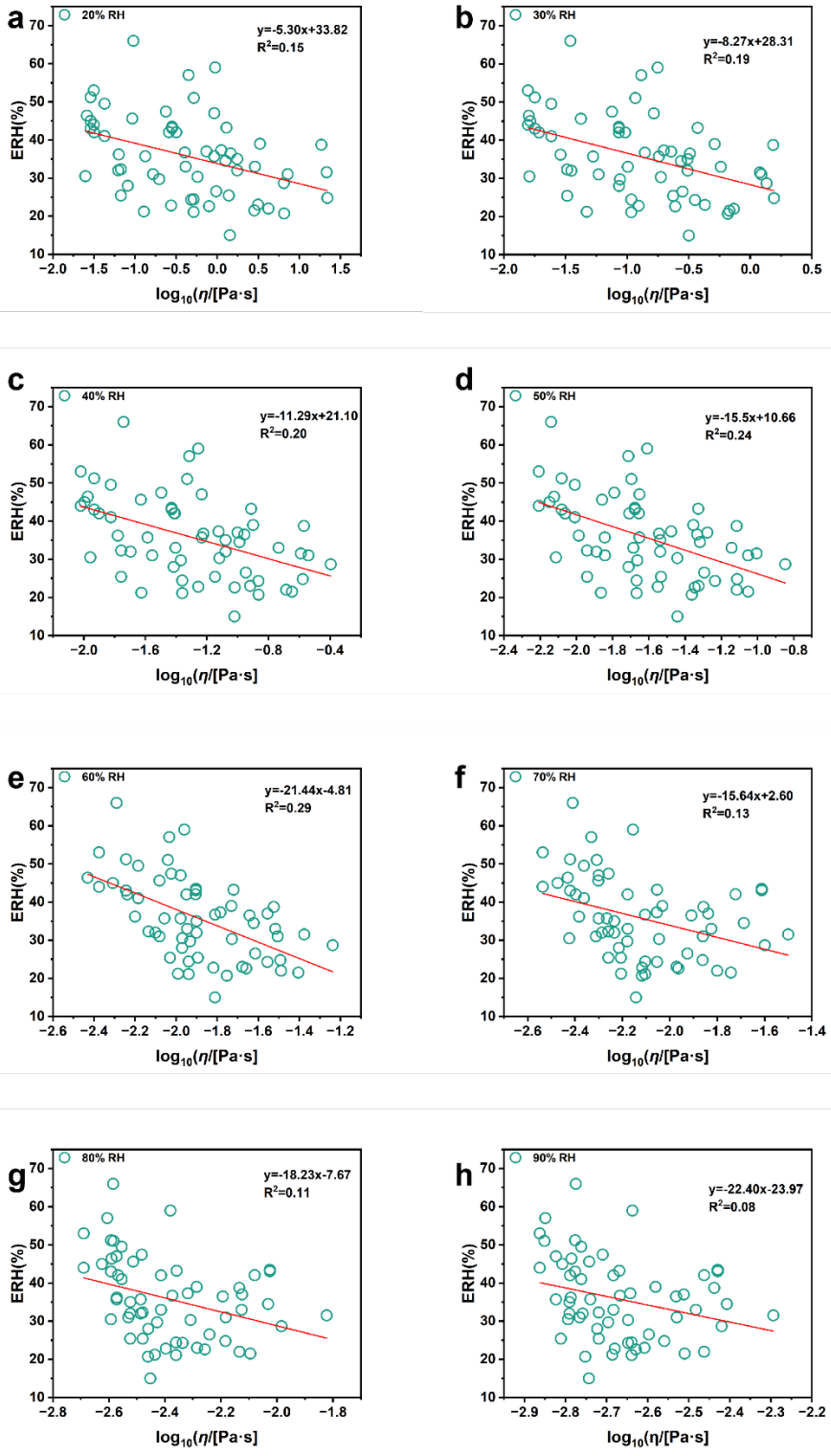


Figure S8. Relationships between ERH and aerosol viscosity calculated at fixed RH conditions. Panels (a–h) show ERH plotted against (log10(η/[Pa·s])), where η represents the aerosol viscosity calculated at fixed RH values of 20%, 30%, 40%, 50%, 60%, 70%, 80%, and 90%, respectively.