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

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

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* $log_{10}v - \frac{\Delta G(T)}{2.303kT}$*. 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”.

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