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

Dear. Dr. Giorio,

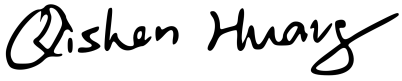
On behalf of my co-authors, I'm pleased to submit a revised version of our manuscript, entitled "Characterizing Efflorescence Regimes in Organic–Inorganic Aerosols Using Thermodynamically Modeled Viscosity" (previously titled "Predicting Organic–Inorganic Aerosol Efflorescence Using Thermodynamically Modeled Viscosity"), for further consideration in *Atmospheric Chemistry and Physics*.

We greatly appreciate the reviewers' positive feedback on our previous revisions. In this round of revision, we have updated the manuscript title and revised figure captions following the reviewers' suggestions. We believe that the new title more accurately reflects the scope and findings of the study, and the revised figure captions further improve the clarity of the manuscript.

A detailed point-by-point response is provided in accompany with this response letter, and all changes are highlighted in the marked manuscript.

We sincerely appreciate the time and effort invested by you and the reviewers in evaluating our manuscript, and we are grateful for your support throughout the review and revision process.

Sincerely,



Qishen Huang

Referee Report 1 (Referee #4):

The authors have addressed my (Referee #4) comments from the last round of reviews adequately. Their other changes to the manuscript, including due to constructive criticism by other reviewers about the effectiveness of using viscosity as causal predictor for efflorescence, have resulted in substantial revisions. These revisions, in my opinion, have strongly reduced the initial premise that viscosity explains or predicts (alone) efflorescence RH, which is good. As a consequence, the revised article has been reframing "the viscosity–ERH relationship as an empirical viscosity–humidity framework defining phase-transition boundaries rather than evidence of a direct mechanistic relationship; …" (see authors' response document).

1) I agree with the authors that the revised article provides now a more balanced assessment of the influencing factors on the efflorescence RH of inorganic aerosol constituents. For that purpose, I find the article and the presented data and analysis insightful. Nonetheless, the intent implied by the original title: "Predicting Organic–Inorganic Aerosol Efflorescence Using Thermodynamically Modeled Viscosity" is no longer consistent with the findings and reframing of the article. I strongly suggest that the title of the article should be revised. In particular, "predicting" may be too strong of a claim now and efflorescence should perhaps be "efflorescence regimes".

Response: We thank the reviewer for the positive evaluation of our revised manuscript. According to the reviewer's suggestion, we have changed the title of the manuscript to ***"Characterizing Efflorescence Regimes in Organic–Inorganic Aerosols Using Thermodynamically Modeled Viscosity"***.

2) Applies to all figures in the main text: please add information about the temperature (range) of the data shown in the figures. This is important especially for the figures/panels showing viscosity, which is strongly temperature-dependent. More generally, ERH itself is temperature-dependent too.

Response: We thank the reviewer for this suggestion. We have added the temperature range of the measured ERH data and the temperature used for AIOMFAC viscosity calculations to all relevant figures in both the main text and the SI.

Revision: Figure 1 – 3 captions: *"All empirical ERH values were measured within a temperature range of 298 ± 10 K, and all viscosity values were calculated at 298 K."* ***Temperature information has also been added to the captions of Figures S2–S4, S7, and S8.***

3) Fig. S7, caption: it is stated "The data points represent the modeled viscosities of different organic–inorganic aerosol compositions at selected RH values 20% to 60%." – by different aerosol compositions, are different solute molar mixing ratios, e.g. 1:1, 1:2, etc. meant or rather that all are e.g. 1:1 but for different solute species in ternary mixtures. Please clarify the phrasing and also state the temperature range for the data shown in the caption.

Response: We thank the reviewer for this suggestion. In Fig. S7, "different organic–inorganic aerosol compositions" refers to all systems listed in Table S1, including variations in both chemical composition and organic-to-inorganic ratio (OIR). We have clarified this in the revised caption of Figure S7 and added details of the representative systems. These representative compositions are also listed in Table S12, which was included in our previous revision.

Revision: Figure S7 caption: *"Relationship between relative humidity (RH) and aerosol viscosity for organic–inorganic mixed aerosol systems. Data points represent modeled viscosities for all aerosol systems listed in Table S1, including variations in both chemical composition and organic-to-inorganic mass ratio (OIR) over an RH range of 20% to 60%. Solid lines show linear fits for four representative systems: Glutaric acid/NaCl (OIR = 0.25, purple), Glycerol/$NaNO_3$ (OIR = 0.25, black), Citric acid/AS (OIR = 0.25, red), and Sucrose/$NaNO_3$ (OIR = 0.25, blue). Details of these representative systems are provided in Table S12. All viscosity values were calculated at 298 K."*