

We thank the referees for carefully reading our manuscript and for their valuable comments. Listed below are our responses in blue to the comments from the referees of our manuscript.

Response to Referee #2

Summary: In this paper the authors quantify the change in viscosity of particles comprised of inorganic salts mixed with sucrose. I think the paper is well written and there is a clear discussion around the experimental procedure and results shown. It is important that more studies quantify the behaviour of mixed inorganic-organic systems and it is refreshing to see a submission focusing on targeted laboratory studies rather than broad extrapolation to global conditions. Therefore, I believe the article can be published in ACP subject to a few general points raised below.

General Comments of Referee #2

[1] Whilst I do fully support the focus on simple mixtures, it would be nice for the authors to provide more context on why these salts were chosen and the expected source of particles with these mass ratios studied. Apologies if I missed this in the document.

[A1] Thank you for the comment. In Sect. 1, we have described the reason why we selected the salts and organic compound as the model compounds as below:

“Sucrose was selected as the model organic substance because previous studies have frequently applied it as a surrogate species for SOA (Zobrist et al., 2011; Power et al., 2013; Grayson et al., 2016b; Song et al., 2016b; Rothfuss and Petters, 2017; Rovelli et al., 2019). Ca(NO₃)₂ and Mg(NO₃)₂ were used as model inorganic salts because they have been commonly observed in mineral dust particles (Usher et al., 2003; Laskin et al., 2005; Sullivan et al., 2007), and sea-salts (Gupta et al., 2015; Zieger et al., 2017) in the atmosphere (Usher et al., 2003; Laskin et al., 2005; Sullivan et al., 2007; Shi et al., 2008; Song et al., 2010, 2013; Pan et al., 2017). Moreover, both of these nitrate salts have a relatively low efflorescence RH in aqueous solutions, enabling viscosity measurements of crystal-free solutions from high RH down to at least 30 % RH.”

[2] Likewise, can the authors comment on the expected timescales for equilibration under ambient conditions? This does not detract from the important of providing more data on previously unstudied systems.

[A2] Based on the work by Koop (2011) and Shiraiwa et al. (2011), the expected timescales for equilibration of various particles ranged from seconds to years depending on the viscosities and materials. Ambient aerosol particles may equilibrate with the gas-phase water vapor within a few seconds to hours or even years. However, further studies are needed to confirm the equilibrating timescales of mixed organic–inorganic particles under ambient conditions, including the timescale for the equilibration of semivolatile organics and inorganics (aside from that of water). Added complexity in particle morphology, such as phase separation, can also influence the equilibration timescale (e.g. Huang et al. 2021); however, our systems did not exhibit phase separation for the studied mixing ratios. To address the Referee’s comment, the following text will be added to the manuscript in Sect. 4.

“Further studies are needed to confirm the equilibrating timescales of mixed organic–inorganic particles under ambient conditions, including the timescale for the equilibration of semivolatile organics and inorganics. Added complexity in particle morphology, such as phase separation, can also influence the equilibration timescale (e.g. Huang et al. 2021); however, our systems did not exhibit phase separation for the studied mixing ratios.”

References:

Huang, Y., Mahrt, F., Xu, S., Shiraiwa, M., Zuend, A. and Bertram, A. K.: Coexistence of three liquid phases in individual atmospheric aerosol particles, *Proc. Natl. Acad. Sci. U. S. A.*, 118(16), e2102512118, doi:<https://doi.org/10.1073/pnas.2102512118>, 2021.

Koop, T., Bookhold, J., Shiraiwa, M., Pöschl, U. and Poeschl, 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(43), 19238–55, doi:[10.1039/c1cp22617g](https://doi.org/10.1039/c1cp22617g), 2011.

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(27), 11003–8, doi:[10.1073/pnas.1103045108](https://doi.org/10.1073/pnas.1103045108), 2011.

[3] Minor comment Page 8, line 224: ‘the shown AIOMFAC-VISC predictions for the ternary systems use an OIR of..’ please change this to ‘the AIOMFAC-VISC..’

[A3] Thank you for the comment. We will revise the manuscript accordingly.