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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Viscosities, diffusion coefficients, and mixing times of intrinsic fluorescent organic molecules in brown limonene secondary organic aerosol and tests of the Stokes-Einstein equation" *by* Dagny A. Ullmann et al.

Anonymous Referee #2

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Summary:

'Viscosities, diffusion coefficients, and mixing times of intrinsic fluorescent organic molecules in brown limonene secondary organic aerosol and tests of the Stokes-Einstein equation' combines viscosity and diffusivity measurements across a range of water activities for brown limonene SOA, and characterizes the accuracy of Stokes-Einstein relation for this system. The brown limonene SOA was generated using dark

ozonolysis of d-limonene and then collected with an impactor and exposed to ammonia.

For the viscosity characterization, the authors used the bead-mobility technique for water activities higher than 0.7. For lower water activities, previously published data using poke-flow techniques by Hinks et al. are used. For the diffusion coefficient and mixing time characterization, a nice aspect of the paper, the authors measure the diffusion coefficient of fluorescent molecules using 'rectangular area fluorescence recovery after photobleaching' (rFRAP). A thin film of brown LSOA was prepared between hydrophobic glass slides. A small area was then photobleached with a laser beam, and the fluorescence is allowed to recover by diffusion of fluorophores into the photobleached region. The measurements in the study were used to test the accuracy of Stokes-Einstein relation, which is commonly used to infer diffusivity from viscosity measurements. It was found that the Stokes-Einstein relationship gave good agreement with measured values over several orders of magnitude in viscosity.

This paper is well-written and the experiments well-executed, with results useful for the community. There are some minor questions/comments listed below that the authors could better address to improve the clarity of the paper. Overall, I recommend publication in ACP.

Specific Comments"

1. What is the chemical identity of the fluorophores (the "intrinsic fluorescent organic molecules") in brown limonene SOA? Are they present in other SOA? Can the diffusivity measurement used here be extended to other systems?

2. Consider very briefly explaining the poke-flow technique for the uninitiated reader, with description of limitations and uncertainties, since the results are used at low water activities.

3. A schematic of the rFRAP technique/set-up, central to this paper, would be appre-

ciated in the SI. Additionally, the SI would be more instructive if it includes details of the thin-film preparation process, with illustrative images of the slides after preparation. Also, can the authors comment on why 2D FRAP was used, instead of the more traditional 1D FRAP? Is there some advantage? If 2D is somehow better, why rectangular, and not circular (for symmetry, which would likely simplify the analysis).

4. Line 178 - Why were the bleach and image sizes chosen based on water activity? Is there a calibration curve for water activity versus area bleached? Does changing the area affect the time of measurement?

5. Line 28 of the abstract contains the abbreviation 'PBL' without first being defined (it is defined later in the introduction. Additionally, the term 'LSOM' has been used in the figures and SI, but LSOA is used in the main manuscript.

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