Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-740-RC2, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diffusion coefficients of organic molecules in sucrose-water solutions and comparison with Stokes–Einstein predictions" *by* Yuri Chenyakin et al.

Anonymous Referee #2

Received and published: 23 September 2016

The manuscript describes a novel method, applied to a problem in atmospheric aerosol science for the first time, to measure the diffusion of organic components with deposited droplets of aqueous solutions of sucrose at varying water activity and, thus, viscosity. Not only is the technique a significant new contribution in its own right to study aerosols, but the measurements that result are extremely useful in contributing to the continuing debate about the diffusion constants of organic species in viscous matrices in secondary organic aerosol. The manuscript is well-written and should be accepted for publication once the authors have responded to the following comments.

- To be consistent with all previous work and avoid confusion, I recommend that all units for diffusion constants be either cm2 s-1 or m2 s-1 throughout the manuscript.

Printer-friendly version

Discussion paper

- The uncertainty in RH of +/- 2.5 % is quite large. Given the steepness in the viscosity vs water activity dependence, significant error must be incurred from this. For conditioning as supersaturated solutions, how long was required for conditioning?

- Page 7: "...independent of the z-direction (i.e. the depth in the thin film), which is a reasonable approximation in our experiments due to the use of thin films (30-50 μm) and the use a 10x objective lens with a low numerical aperture, which gives a near cylindrical geometry over a distance of 30-50 μm in the z-direct." It is surprising to me that a 10x objective gives such a large Rayleigh range in the beam waist. I would have expected the beam to diverge more quickly than this beyond the focal waist and for the shape to not be of simple cylindrical geometry. Can the authors back up this claim with some actual calculations for their particular system?

- Photobleaching recovery: Following on from the last point about the geometry of the bleached volume, it must be presumed that there is no recovery following photobleaching apart from that due to diffusion of organic species back into the detection window of this assumed geometry. It would be helpful to confirm this by carrying out photobleaching recovery experiments at very low relative humidity in sucrose where the diffusion is most likely entirely quenched. Have the authors done this?

- Hydrodynamic radius of fluorescein: How confident can the authors be that the hydrodynamic radius is independent of water activity? This would seem to be quite crucial when considering the plausibility of the Stokes-Einstein equation for reproducing the diffusion constants from measured viscosities. Given that many of the estimated diffusion constants overlap with the measured diffusion constants, it seems possible to me that Stokes-Einstein could even be considered to be a good representation of the diffusion constants of the organic dyes, given all of the uncertainties/errors involved (including the uncertainties in RH and the uncertainties in the viscosity measurements of sucrose solutions).

- Given the disparity in sizes of the different fluorescent probes, would the authors

Printer-friendly version

Discussion paper

expect any of them to be better represented by Stokes-Einstein behavior, once their sizes are compared with that of sucrose?

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-740, 2016.

Printer-friendly version

Discussion paper