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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Viscous organic aerosol particles in the upper troposphere: diffusivity-controlled water uptake and ice nucleation?" *by* D. M. Lienhard et al.

Anonymous Referee #2

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This paper reports very interesting findings on the ability to generalise diffusion coefficients and hypothesise potential impacts. The paper provides a refreshing investigation in a field that already has a growing number of reports in high impact journals. I believe the paper should certainly be published once the authors have considered and responded to the following:

Page 24478: line: 'The numerical model subdivides the particle into up to several thousand individual shells and solves the non-linear diffusion equation in spherical coordinates while accounting for the concentration dependence of the water diffusion coefficient, i.e. accounting for the plasticizing effect of water . . .'. I'm curious as to

what this means, physically, when say a 900nm particle is split into a few thousands shells? I presume this has been covered in previous papers but what effects do a 'sub molecular' representation mean for modelling the diffusion process? Is it because the numerics indicate a tangential behaviour for equilibration times when resolution is continually increased?

On the same page:'the diffusion coefficients of the solutes are expected to be much slower than water and are not accounted for.' Does this mean the solute is assumed not to diffuse with water? In other words, if you assume a system with symmetric diffusion coefficients, this would effectively result from assuming an ideal binary system within a Fickian framework. If you are not assuming this, presumably non-ideality is accounted for? I guess the easier way to answer this, is, what is the assumed diffusion coefficient for the solute with significant amounts of water?

Section 3. Do the authors expect any semi-volatile loss from the alpha-pinene aerosol when extracting diffusion coefficients of water? I'm just curious as to the use of these inferred diffusion coefficients in an atmospheric simulation somehow has an inherent effect from such a process?

In the abstract the authors note that: 'condensed-phase water diffusivity is unlikely to have significant consequences on the direct climatic effects of SOA particles under tropospheric conditions.' In the general atmospheric simulations, it isn't clear in the text whether size distribution dynamics are accounted for. Despite the hypothesis that, at the single particle level, time-scales are significantly reduced to remove the 'importance' of diffusion, isn't there a potential effect on size distributions from a non-instantaneous equilibration below cloud? Is this accounted for? I would expect a parcel model with the diffusion model accounted for to be quite expensive, perhaps I am wrong. The results from this model would also likely be sensitive to a range of initialisation conditions including assumed history of water uptake, size distribution, inorganic core and up draft?

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