

Interactive comment on “The Rate of Equilibration of Viscous Aerosol Particles” by S. O’Meara et al.

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We thank the reviewer for their helpful comments. We have included our response in the supplementary material.

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We thank the reviewer for their helpful comments. All comments were thought beneficial to our study and we have described the resulting modifications to the paper below. Reviewer comment are numbered and italicised while our responses are given below each comment and indented.

1) In the last paragraph of the introduction the authors mention that a Fickian framework may not be appropriate for some systems, in particular glassy polymers. While an extensive discussion is clearly beyond the scope of the manuscript, it will help the reader if the authors give some basic information in a few sentences on what causes non-Fickian diffusion, i.e. mechanical deformation in response of a solvent diffusing in a matrix.

A brief description of the process leading to divergence from Fickian diffusion has been included. p 2 line 5 now reads:

* Non-Fickian diffusion results from structural changes following diffusion and the resultant composition change. It arises when the rate of deformation is comparable to that of diffusion (Crank 1975).*

2) Discussion of Fig. 1b and the figure: For a reader not familiar with the topic it would be helpful to introduce first the case of Fickian diffusion with a concentration independent diffusion constant and only then compare to one in which the diffusing species acts as a plasticizer. Also, the caption of Fig. 1b should contain the info that it shows the response to an instantaneous increase in RH from 10 to 20%.

A further subplot was added to Figure 1 to illustrate the differences in concentration-radius profiles for different dependencies of the diffusion coefficient on concentration. The associated discussion starting at pp. 3 line 21 now reads:

Figure 1a demonstrates how the particle is represented in a 2-D view. Fig. 1b illustrates the concentration-radius profiles of a semi-volatile component at several time steps using the ETH model in the case of an instantaneous increase in saturation ratio from 1 to 90% when the diffusion coefficient is independent of composition. In contrast, Fig. 1c shows the same information but when the diffusion coefficient has a logarithmic dependence on composition and the self-diffusion coefficients of the two components are very different, that of the non-volatile ($D_{ns}^0 = 1 \times 10^{-21} \text{ m}^2 \text{ s}^{-1}$) and for the semi-volatile ($D_{sv}^0 = 2 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$). The “diffusion front” is clear in this example and arises from the very different diffusion coefficient values in neighbouring shells that result from variations in shell composition.

3) The authors use τ -folding times as a metric for diffusion time. Of course this is technically correct, but may lead to a misunderstanding for a reader who is not familiar with the topic. The temporal response of system in which the diffusing species acts as a plasticizer cannot be described by a single exponential. The approach of the authors to take as a measure the time when the difference between average bulk and surface concentration has changed by e is valid, but a short discussion is appropriate.

This point is welcome and has motivated extra discussion in the method, pp. 4 line 6 now reads:

*Comparing τ -folding times between models strictly only tests model consistency at this particular stage of diffusion and not before this. However, τ -folding time agreement would indicate agreement at previous times (and future ones), because the

Fig. 1.

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