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_{nv}^{0}) = 1x10^{-21} m^{2}s^{-1} and for the semi-volatile (D_{sv}^{0}) = 2x10-9 m^{2}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 e-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 *e*-folding times between models strictly only tests model consistency at this particular stage of diffusion and not before this. However, *e*-folding time agreement would indicate agreement at previous times (and future ones), because the

underlying equations are identical. For reassurance on this, concentration-radius profiles at times prior to e-folding were compared."

4) In the same context: I would very much appreciate at least one example in which the authors do not only compare e-folding times (in the sense mentioned above), but directly compare calculated profiles with the three models for a case leading to a steep diffusion profile. Did they observe any differences between the models here?

Providing the example described benefits the evaluation of model consistency. The modifications to the paper described below are associated with the modification described in reviewer point 3). A new figure (Fig. 5) has been included to present the agreement between models for concentration-radius profiles.

pp. 10 line 15 now reads:

" As discussed, the agreement between models in estimating e-folding times indicates that the estimated profiles of concentration with particle radius prior to the e-folding state are consistent between models because the underlying equations are the same. By comparing concentration-radius profiles at various stages of diffusion we indeed found good model agreement across all cases. In Fig. 5 we show the example of the logarithmic dependence of D_i on x_{sv}, an instantaneous change in saturation ratio of 1-90% and with D_{nv}^0=1x10^{-21} m^{2}s^{-1} and D_{sv}^0=2x10^{-9} m^{2}s^{-1}. At several times preceding and including e-folding time the concentration-radius profiles are in good agreement."

and pp. 11 line 19 reads:

" The consistency in modelled concentration-radius profiles at times preceding and including the e-folding state (Fig. 5) shows that if used for a polydisperse aerosol population, the models would give agreement in changes to the size distribution. In addition, if the diffusing component were reactive the rate of particle-phase reaction would depend on its concentration; therefore model agreement in concentration-radius profiles would give consistent reaction rates across the particle (which in turn could affect diffusion rate)."

5) *And last: there is no discussion on how the models compare in terms of computational speed. Of course this my dependent on the specific coding and comparison may not be easy, but either there is a significant difference between the ones coded by the authors or not. Whatever is the answer, it is of interest for a reader who would like to use one of the model framework*

This additional information we agree would be beneficial to readers, so we have included the following illustration of computational speed variations in pp. 9 line 4:

"Using the Matlab software it was found that computational time for the case of diffusion coefficient independent of composition was quickest, gradually increasing as the steepness of the diffusion coefficient dependence on composition increased, largely due to the greater spatial resolution. For D_i independent of composition the ETH model took of the order 1 s to reach the e-folding state while KM-GAP and Fi-PaD were of the order 10^2 s. For a steep diffusion coefficient dependence, the chosen example was the logarithmic dependence, with D_{nv}^0=1x10^{-25} m^{2}s^{-1} and D_{sv}^0=2x10^{-9} m^{2}s^{-1} and e_s instantaneously increased from 1-90%: the ETH model took of the order 10^2 s while both KM-GAP and Fi-PaD took of the order 10^4 s. "

Furthermore, we have a paragraph to the discussion and conclusion, pp. 11 line 25 now says:

" Using the three diffusion models as described above and with the spatial resolutions presented in the appendix, the ETH model takes approximately two orders of magnitude less computer time than Fi-PaD or KM-GAP for a given diffusion scenario. With the models giving consistent estimates of diffusion, the ETH model therefore appears to be favourable."

6) Technical: The Lienhard et al. 2015 paper is published now in ACP.

This reference has been modified accordingly, pp. 14 line 17 now reads:

" Lienhard, D. M., Huisman, A. J., Krieger, U. K., Rudich, Y., Marcolli, C., Luo, B. P., Bones, D. L., Reid, J. P., Lambe, A. T., Canagaratna, M. R., Davidovits, P., Onasch, T. B., Worsnop, D. R., Steimer, S. S., Koop, T., and Peter, T.: Viscous organic aerosol particles in the upper troposphere: diffusivity-controlled water uptake and ice nucleation?, Atmos. Chem. Phys., 15, 13599-13613, doi:10.5194/acpd-15-13599-2015, 2015."