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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Timescales of Secondary Organic Aerosols to Reach Equilibrium at Various Temperatures and Relative Humidities" *by* Ying Li and Manabu Shiraiwa

Anonymous Referee #1

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This manuscript explores the equilibrium timescale and mixing timescale of IVOC and LVOC with particles considering different phase states. The work combines the authors' previous KM-GAP model (Shiraiwa et al., 2012) with the authors' recent glass transition model (Shiraiwa et al., 2017; DeRieux et al. 2018) to understand the interplay among equilibrium timescale, temperature, relative humidity, and the glass transition temperature of the aerosols.

Besides the numerical results obtained from the model shown in Figure 1-5, the manuscript provides two more useful results: (1) when there is no diffusion limitation within the particle, the gases that have higher saturation mass concentrations will reach gas-particle equilibrium faster (2) when there is strong diffusion limitation within the particle, gases that have lower volatility may reach gas-particle equilibrium (locally) faster than VOCs with higher volatility. One of the implications is that at a lower temperature (upper troposphere) or when dealing with highly viscous particles, the particle growth process may need to be treated kinetically.

The authors used a numerical model to obtain result #1 above, and it agrees with the findings in Liu et al. 2012 in which an analytical model was used. It is nice to see two different approaches come with the same results and can validate each other. On the other hand, result #2 is more interesting because it shows that local equilibrium could be reached relatively faster for low volatility species when the particle phase is highly viscous. The manuscript also illustrates some concepts that can be commonly misused by folks, such as the difference between equilibrium timescale and the mixing timescale.

Because some of the results have been previously discussed in or can be easily inferred from other publications (Liu et al. 2012, Shiraiwa et al. 2011&2012), the novelty of the manuscript needs to be improved. I suggest that the author focus on result #2, which is novel, and use it to make further predictions regarding the physical and chemical processes of aerosols. For instance, what is the relationship between particle sizes and condensation/evaporation kinetics of gases with different volatility interacting with particles with various viscosity? I would also be curious to know what is the boundary criteria for result #2 to happen, i.e., how viscous would the particles have to be or how low the volatility of the VOC have to be in order to reach relatively fast local equilibrium? The author can also explore the effects of equilibrium partitioning when the gases can both partition in and react with the particle phase.

My other question is that most of the modeling results shown the manuscript assumed that the gas-particle is in a closed system. How realistic is the closed system in ambient environment? Would the ambient environment often be an open system for evaporation kinetics?

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Discussion paper

The author should also include Price et al. 2015 in the reference list in line 115.

Reference: Shiraiwa et al. 2011: doi.org/10.1073/pnas.1103045108 Liu et al. 2012: doi.org/10.1080/02786826.2012.730163 Shiraiwa et al. 2012: doi.org/10.1029/2012GL054008 Price et al. 2015: doi.org/10.1039/C5SC00685F

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