

Understanding the microstructure of supersaturated organic aerosol

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Extracting macroscopic water kinetics from ultraviscous aerosol measurements (in, for example, optically tweezed droplets)¹ relies on assuming both the form for the diffusion coefficient dependence on activity and radial concentration gradient simultaneously.² Classical Molecular Dynamics (MD) methods offer a novel method for extraction of water diffusivity by modelling portions of an aerosol particle surface small enough that water concentrations remain roughly constant (~6nm³), bypassing both of these limitations.

~10¹ nanosecond simulations are conducted of water in sucrose, a commonly used proxy for atmospheric secondary organic matter, at a variety of supersaturations.

It is found that molecular D_{water} values remain of the same order as pure water at activities down to 0.5, due to diffusion only occurring within the initially occupied volume of water. Newly reported analytical methods³ are applied for the estimation of effective water diffusion coefficients that may more closely relate to macroscopically observed timescales for water transport in atmospheric aerosol particles. Specifically, we consider two regimes: diffusion controlled by motion of clusters isolated from one another, and diffusion within interlinking channels that may be considered to be static. The validity of the input coordinates is assessed by enhanced sampling⁴ of the angular tetrahedrality phase space of the system.

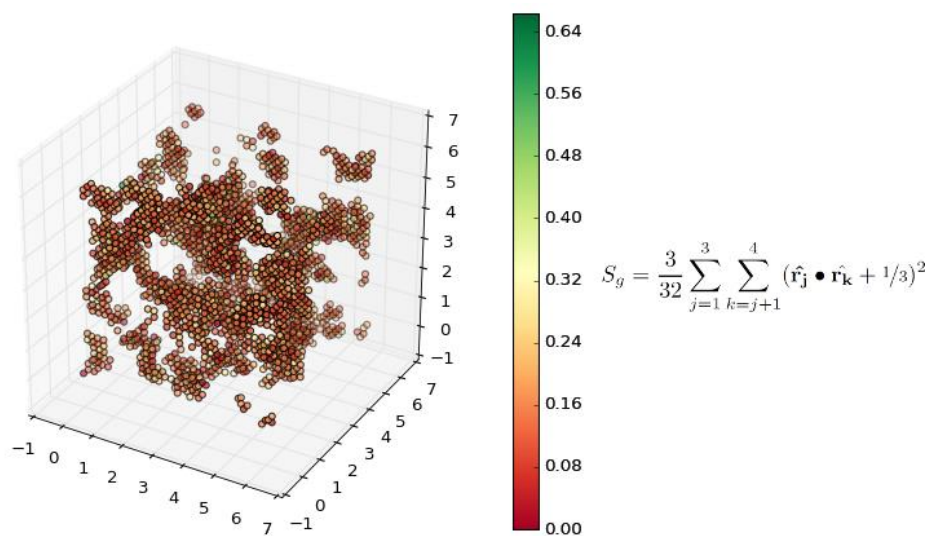


Figure 1: Spatial distribution of the S_g tetrahedrality parameter within the occupied water volume (each point = one molecule) of an 89wt% sucrose simulation box.

References

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