Measurements of Diffusion Coefficients of Organic Dyes in Proxies of Atmospheric Particles and Comparison with Predictions Using the Stokes-Einstein Equation

D. A. Ullmann[1], Y. Chenyakin[1], S. Kamal[1], A. K. Bertram[1]
[1] The Univ. of British Columbia, Vancouver, Canada

Motivation

Why? Particles consisting of secondary organic material (SOM) influence the Earth's climate.[1] To better understand the effect of SOM particles on the Earth's climate, several characteristics of SOM particles such as viscosity have been studied.

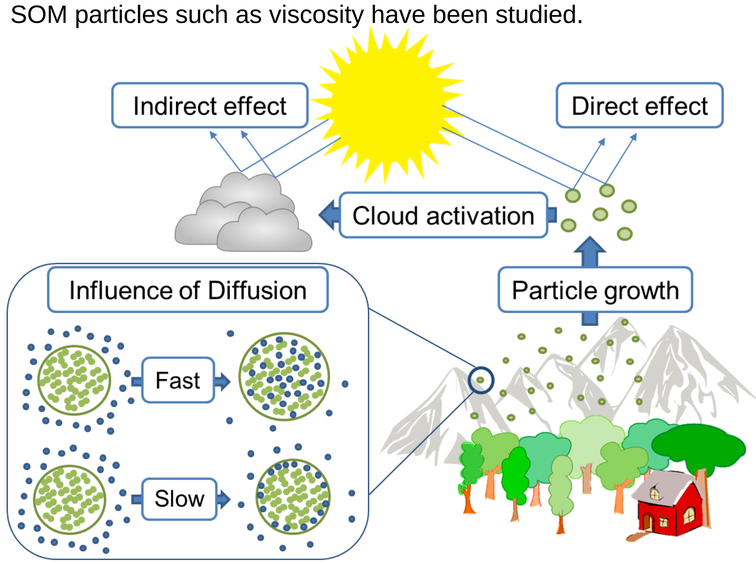

Figure 1: Schematic representation of the formation of atmospheric particles, their effect on climate and the effect of viscosity on particle properties. [1]

Goal: In the past, diffusion coefficients have mostly been calculated using the Stokes-Einstein equation.[2] Here, we present diffusion coefficients which have been measured directly using FRAP (fluorescence recovery after photobleaching).

What is FRAP?

How? Diffusion coefficients can be measured directly by using the FRAP microscopy method. In FRAP, a defined area of a film of a sample containing fluorescent molecules is photobleached using high laser intensity. Due to diffusion of fluorescing molecules within the film, the photobleached molecules diffuse out of the photo-bleached area and get exchanged by intact molecules (fig. 2). [3]

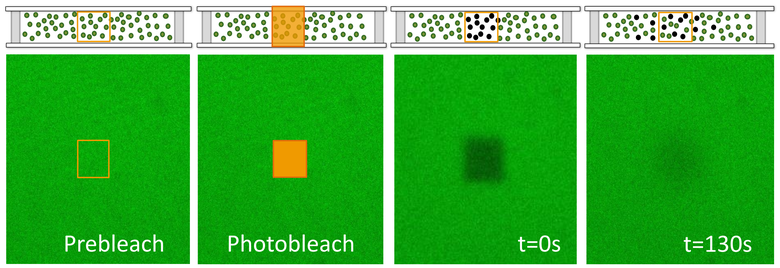

Figure 2: Schematic representation (top) and images of the recovery of a photobleached area (bottom). The photobleached area recovers, as the photoblea-ched molecules (black) get exchanged by intact molecules (green).

Sample Preparation

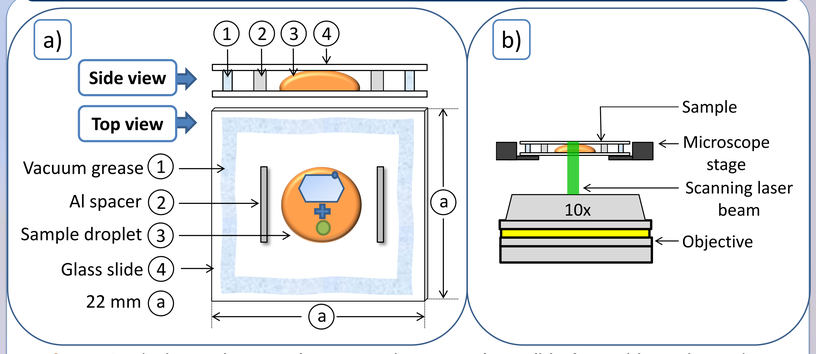

Figure 3: a) shows the sample preparation on a glass slide from side and top view. The sample droplet contains sucrose (blue hexagon) and different fluorescent molecules (green circle; see table 1). b) shows the microscope setup.

Results I - *Diffusion coefficients in proxies of atmospheric particles*

Sucrose-water solutions are known to have similar viscosities to atmospheric particles over a wide range of relative humidities (RH).[4] Sucrose-water solutions containing various fluorescent dyes were prepared and the diffusion coefficients were measured over a wide range of RHs.

Table 1: Fluorescent molecules studied in this project; r_H=Hydrodynamic radius.

Dyes	M_W (g/mol)	r_H (Å)	References r_H
Fluorescein	376	5.02	Mustafa et al. (1993)
Rhodamine 6G	479	5.89	Müller et al. (2008)
Calcein	622	7.4	Tamba et al. (2010)
FITC-Dextran 150	$1.50 \cdot 10^5$	83.1	Smedt et al. (1994)

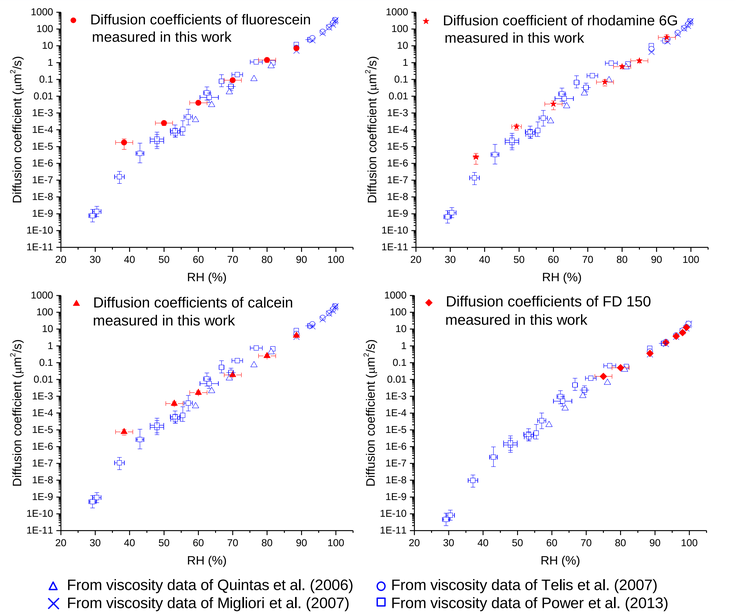

Figure 4: Comparison of the measured diffusion coefficients of the studied dyes with values of diffusion coefficients that were calculated from viscosity data using the Stokes-Einstein equation.

Results II – *Diffusion coefficients in SOM*

Limonene based SOM show fluorescence in the visible light range and it has been shown that it is possible to photobleach these particles.[5] These properties have been used to directly measure diffusion coefficients in SOM at different relative humidities (RHs).

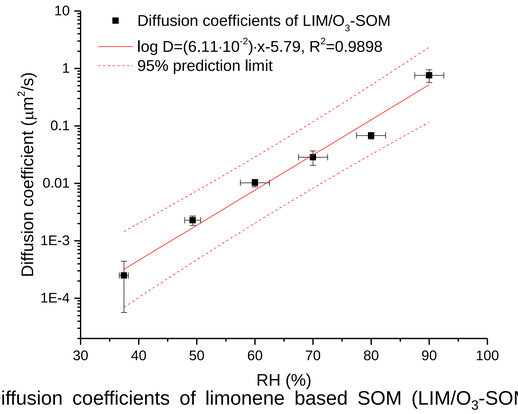

Figure 5: Diffusion coefficients of limonene based SOM (LIM/O_3-SOM) at various RHs.

Conclusions

1) At low RHs the Stokes-Einstein equation breaks down. A correction will be needed to predict diffusion coefficients at low RHs.
2) The diffusion coefficients of the fluorescent molecules in LIM/O_3-SOM do not vary as much with RH as the diffusion coefficients of other fluorescent molecules in sucrose-water particles.

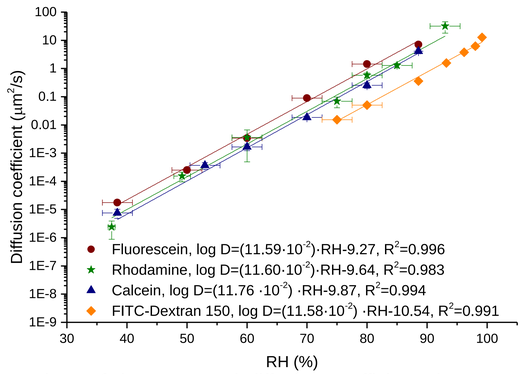

Figure 6: Comparison of the measured diffusion coefficients in sucrose-water particles.

References

[1] S. M. King, T. Rosenoern *et al.*, *Atmospheric Chemistry and Physics,* **2009**, *9*, 2959-2971.
[2] L. Renbaum-Wolff *et al.*, *Proceedings of the National Academy of Sciences,* **2013**, *110*, 8014-8019.
[3] H. Deschout *et al.*, *Opt. Express,* **2010**, *18*, 22886-22905.
[4] B. Zobrist *et al.*, *Atmospheric Chemistry and Physics,* **2008**, *8*, 5221-5244.
[5] D. L. Bones *et al.*, *Journal of Geophysical Research: Atmospheres (1984– 2012),* **2010**, *115*.

Acknowledgements

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC) and by the LASIR laboratory of The University of British Columbia, Vancouver.