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Impacts of Phase State and Water Content on Secondary Organic Aerosol Formation and Partitioning

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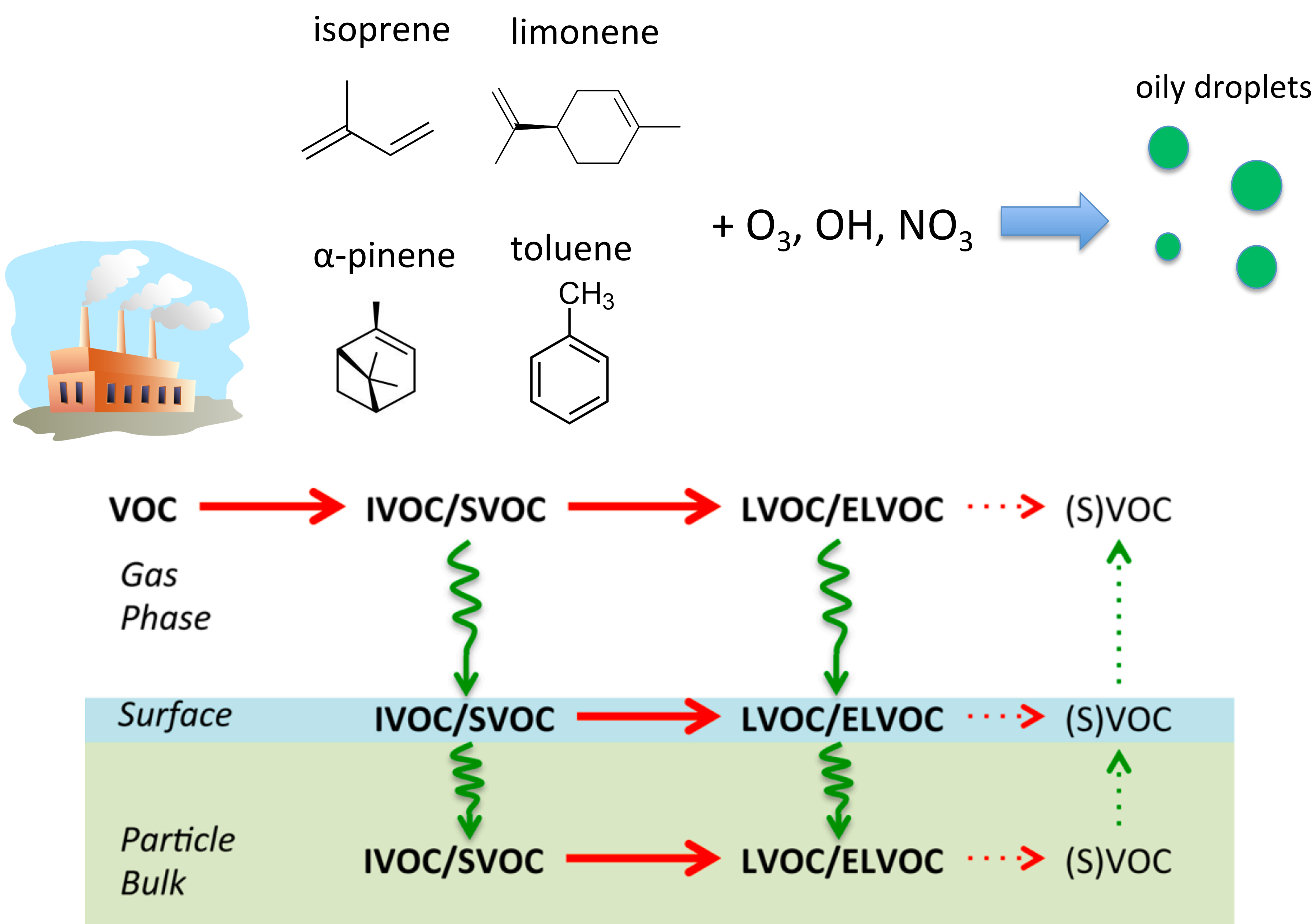
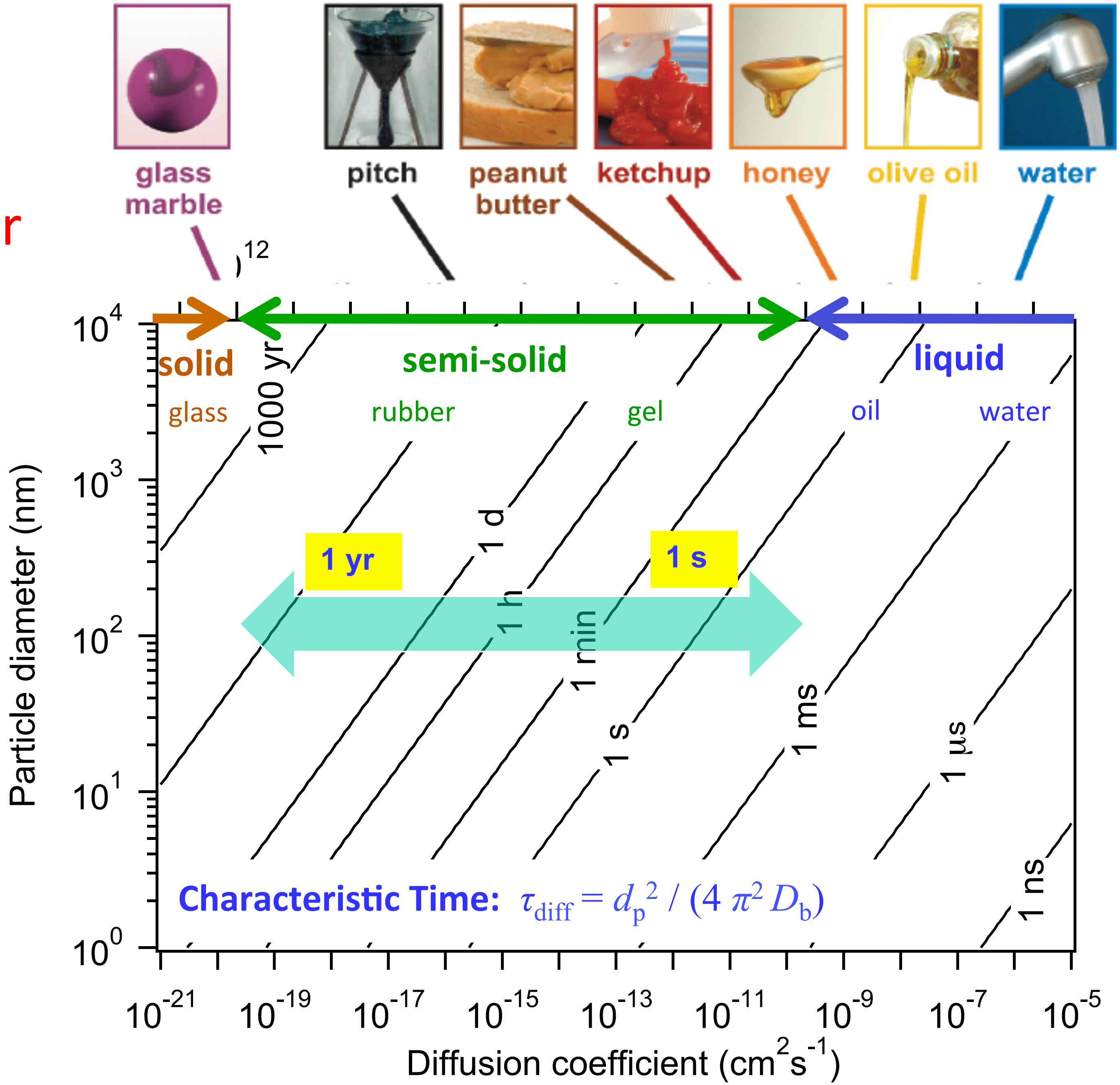
Secondary Organic Aerosol (SOA) Formation



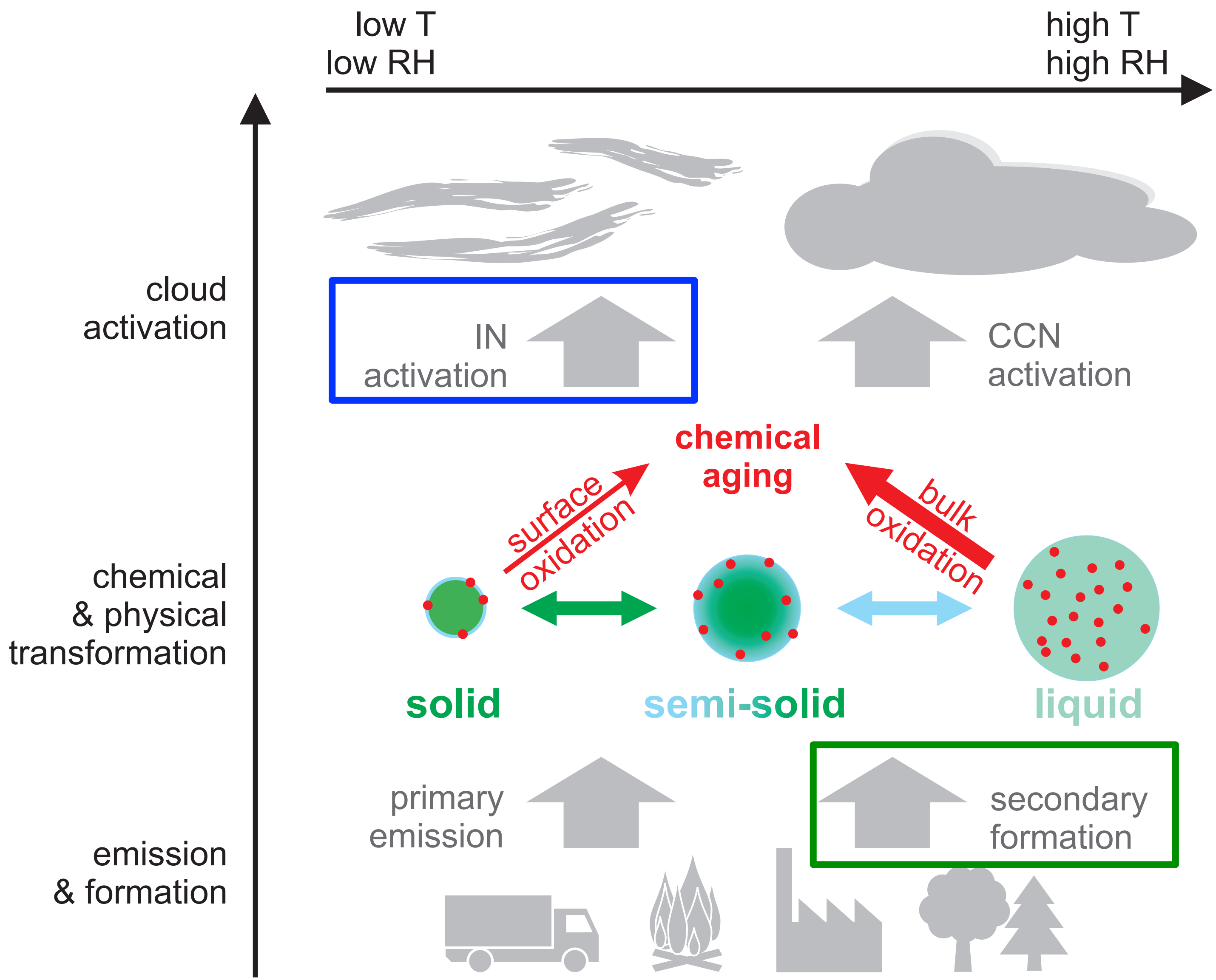
SOA can adopt semi-solid state (highly viscous or glassy)

Zobrist 2008; Mikhailov 2009; Virtanen 2010; Koop 2011; Vaden 2011; Cappa & Wilson 2012; Renbaum-Wolff 2013; Zhang 2015; Song 2015, 2016; Rothfuss & Petters 2017, …



Shiraiwa et al., PNAS, 2011

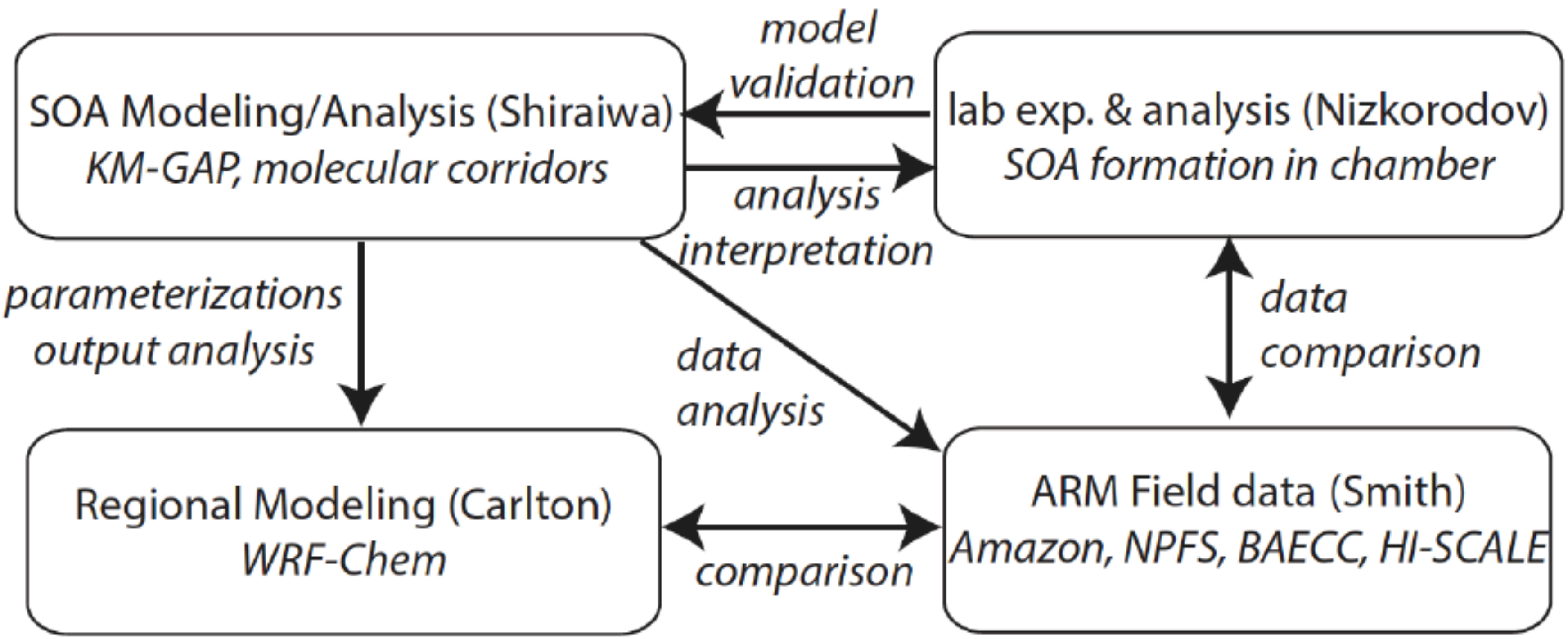
Atmospheric Implications



Perraud 2012; Wang, Knopf 2012; Kuwata & Martin, 2012; Zhou 2013; Kidd 2014; Lignell 2014; Davies 2015; Liu 2016; Slade 2017; Shrivastava 2017; Ye 2018; Zaveri 2018, …

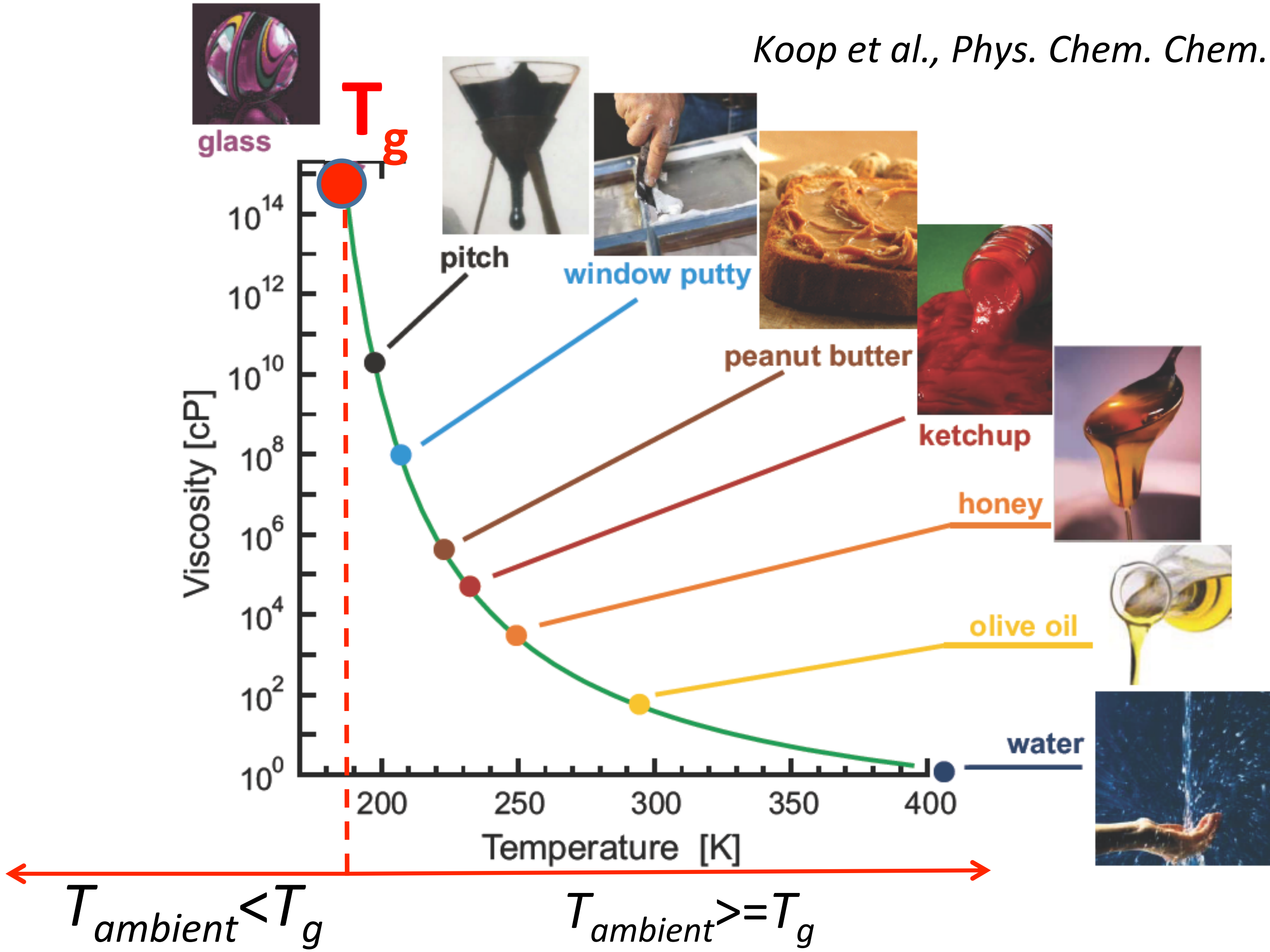
Shiraiwa et al., PNAS, 2011

Phase-Aerosol-Water (PAW) Project (2017 – 2020)



Prediction method: Glass Transition Temperature (T_g)

Koop et al., Phys. Chem. Chem. Phys., 2011



Parameterization to predict T_g using elemental composition (molar mass and O:C ratio, or number of C, H, O) was developed

Shiraiwa, Li et al., Nature Commun., 2017; DeRieux, Li et al., ACPD., 2017

Viscosity Estimation using HR-MS data of Toluene SOA

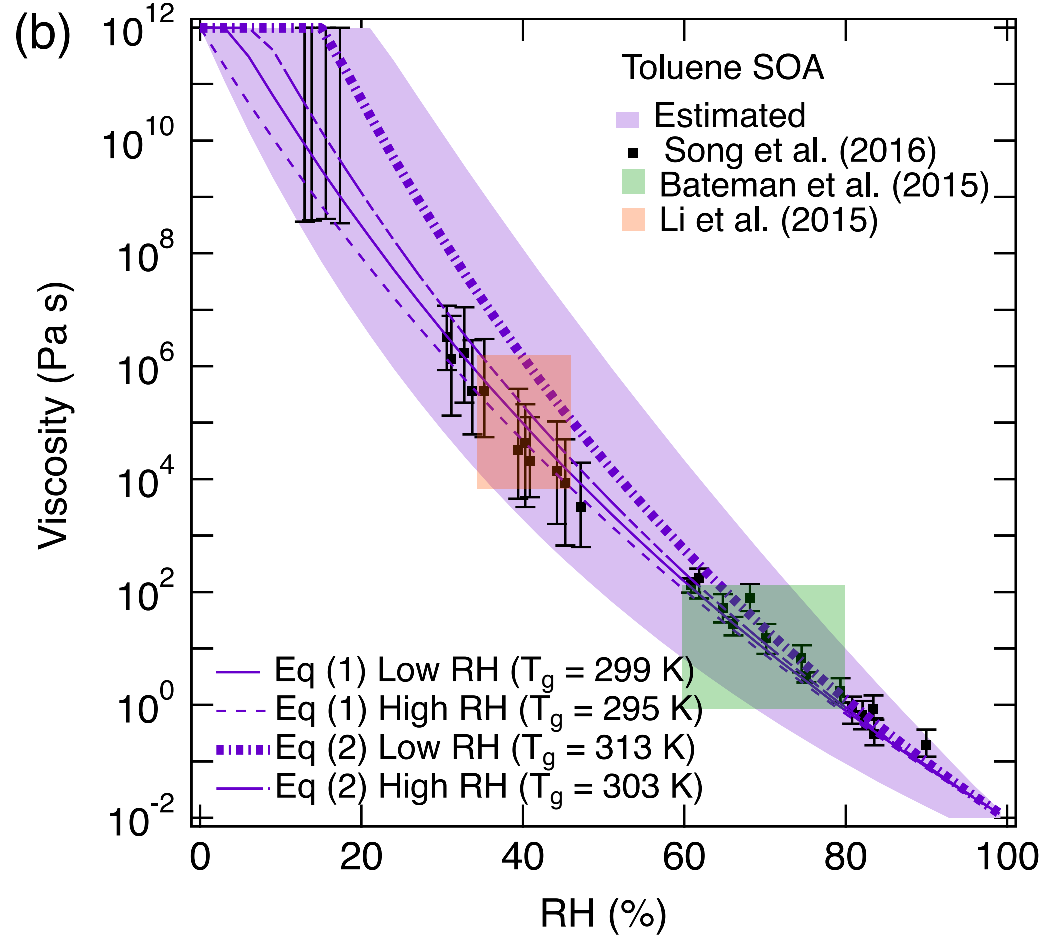
Molecular composition (*Hinks et al., ACP, 2018*)

Prediction of T_g of each compound

Gordon-Taylor equation to estimate T_g of dry SOA

Estimation of T_g of SOA with water by considering hygroscopic growth (κ = 0.2) and the Gordon-Taylor rule (k_{GT} = 2.5)

Estimation of viscosity using the Angell plot of viscosity vs. T_g/T

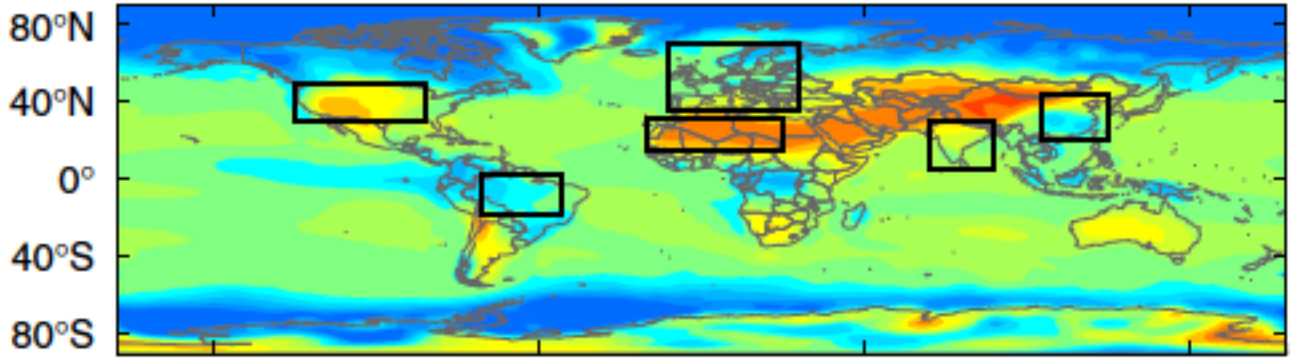


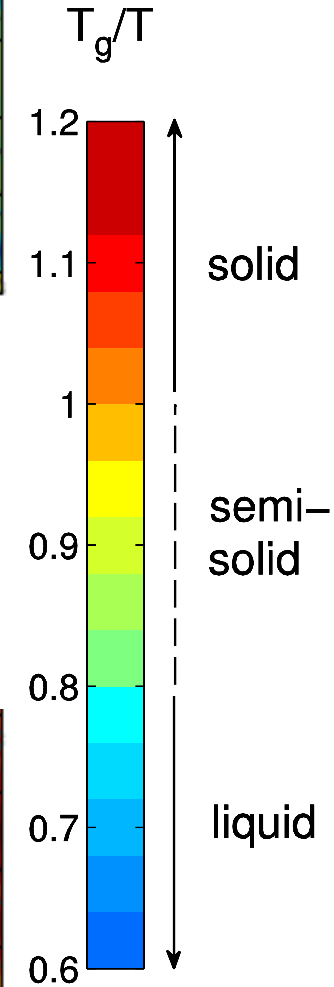
DeRieux, Li et al., Atmos. Chem. Phys. Discuss., 2017

Global Distribution of SOA Phase State of SOA

T_g prediction method combined with a global model EMAC-ORACLE

Surface





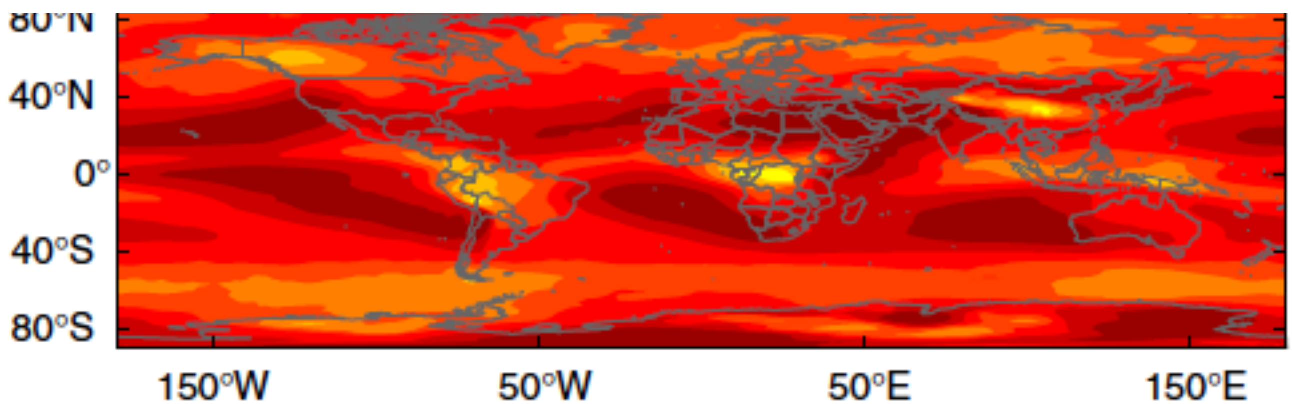
Consistent with field observations:

Liquid in Amazon (Bateman et al., Nat. Geosci., 2015)

Semisolid in Chile, Mexico, California (O’Brien et al., GRL, 2014)

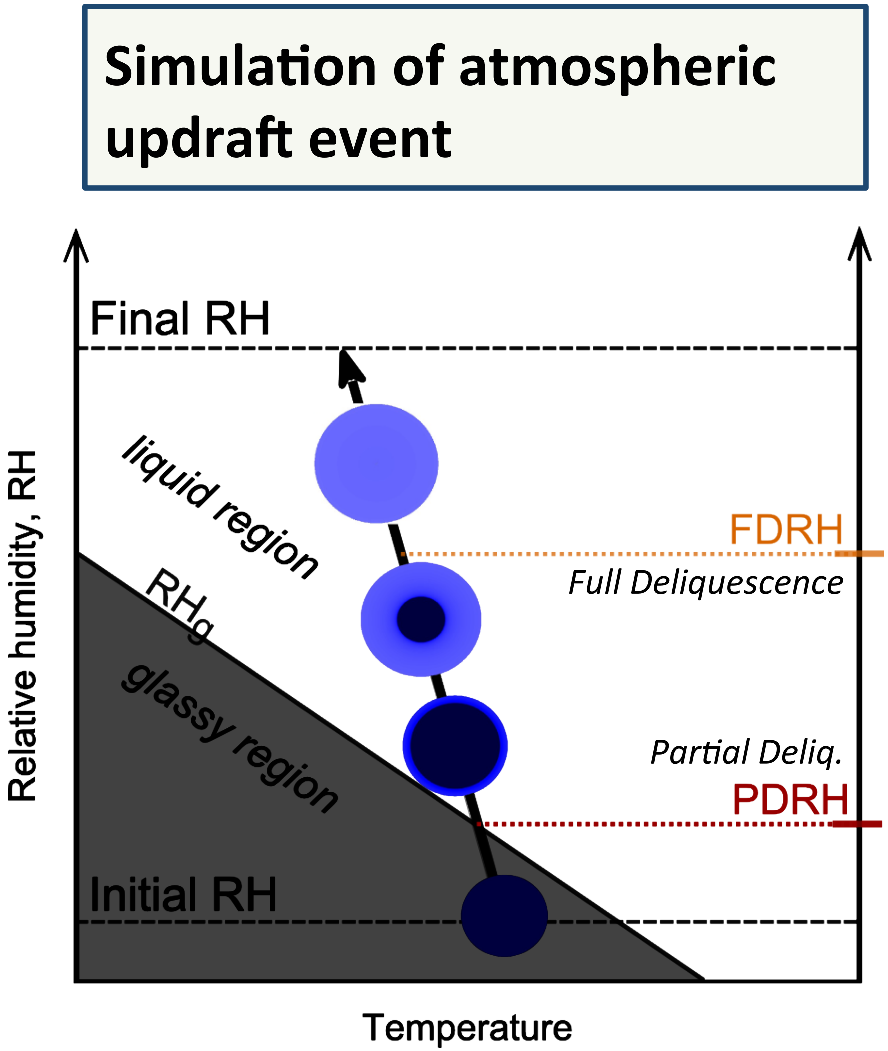
Solid in Finland (Virtanen et al., Nature, 2010)

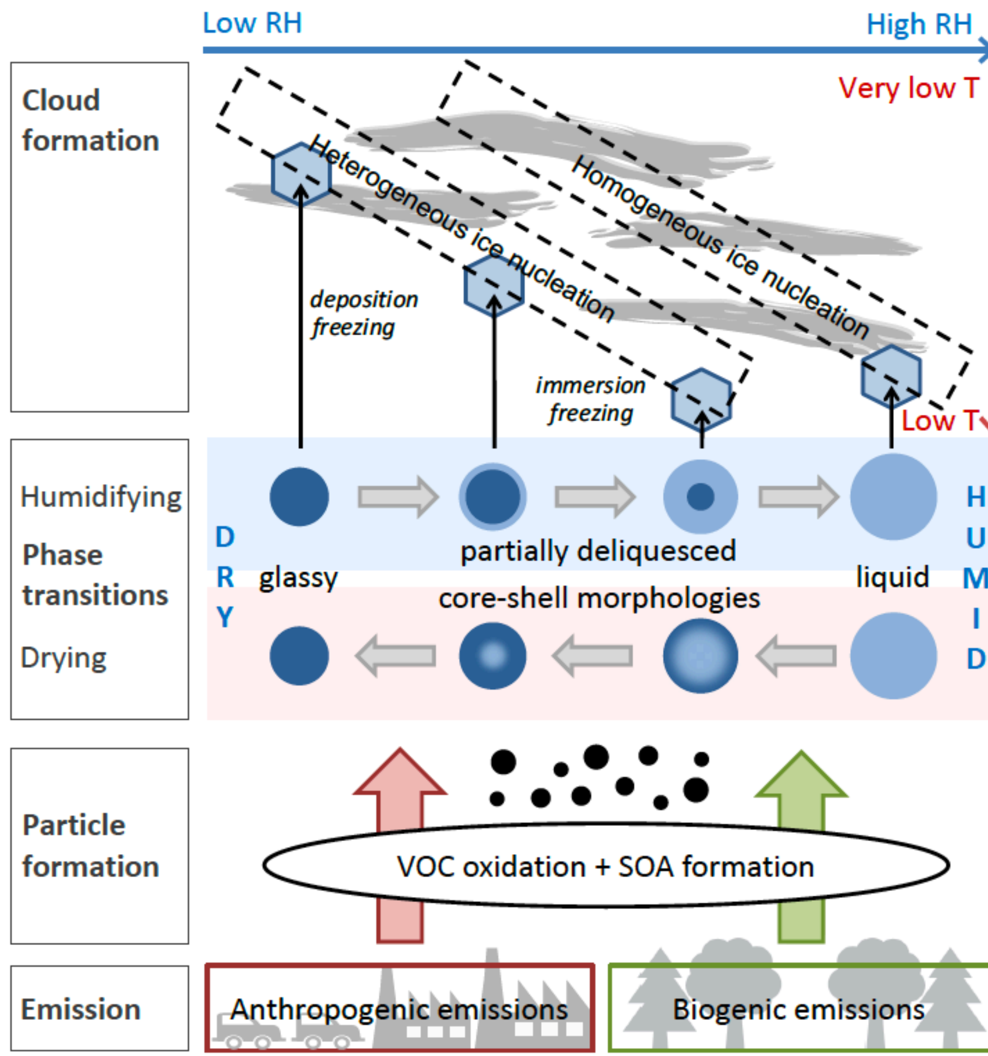
500 hPa



Shiraiwa et al., Nature Commun., 2017

Deliquescence vs. Ice Nucleation

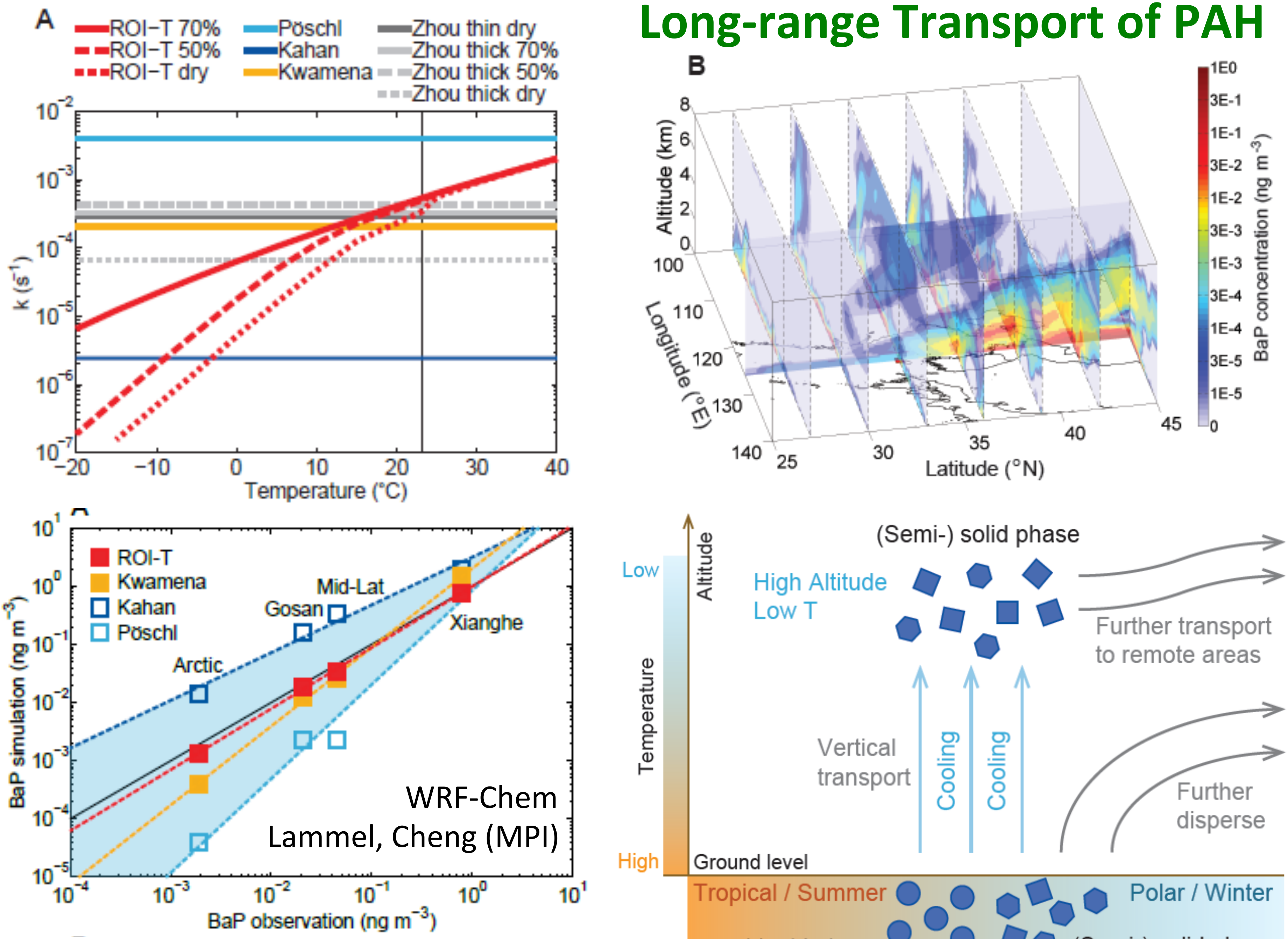




Berkemeier et al., Atmos. Chem. Phys., 2014

Charnawska, Knopf et al., Faraday Discuss, 2017

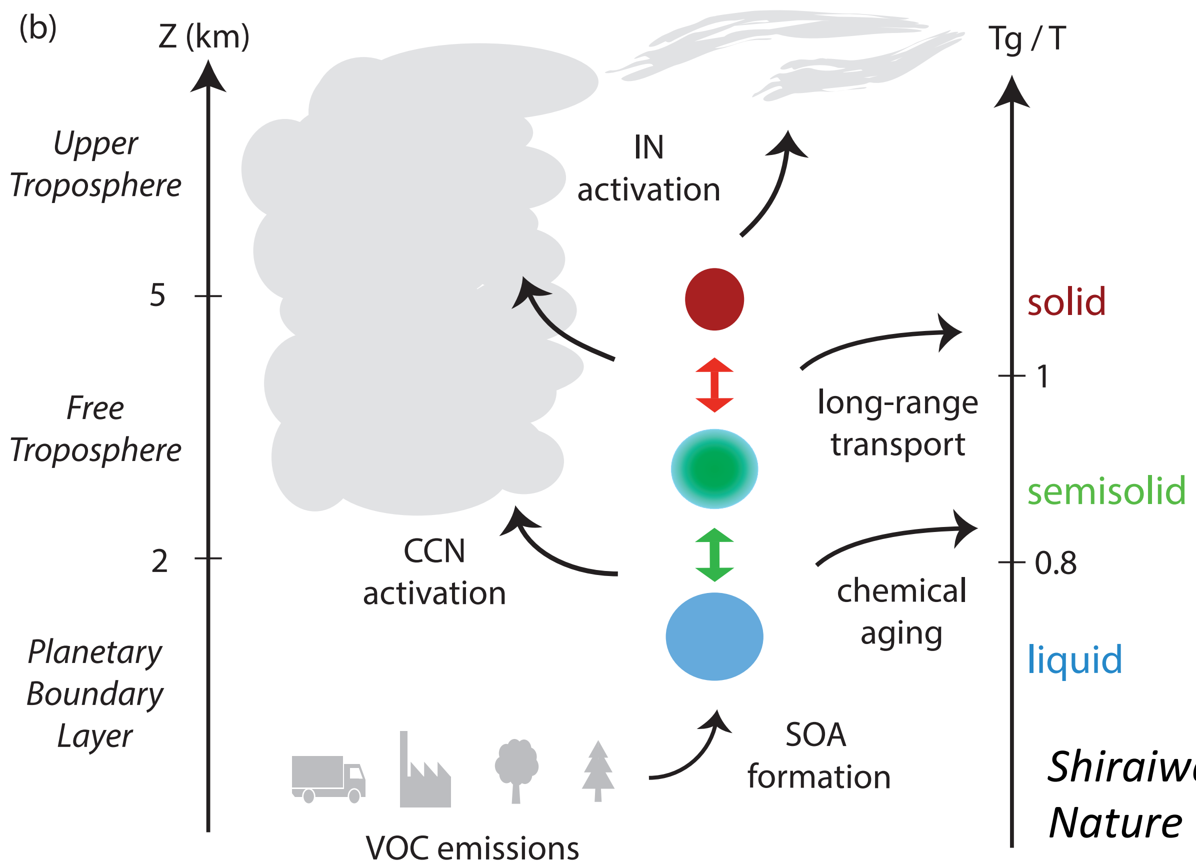
Long-range Transport of PAH



Mu, Shiraiwa et al., Science Advances, 2018

Summary

Old: Organic aerosol particles are liquid oily droplets



Shiraiwa et al., Nature Commun., 2017

New: SOA can be glassy or amorphous semi-solid under low RH & T:

1) facilitating long-range transport of toxic organic pollutants
2) affecting ice nucleation pathways
3) causing kinetic limitations in gas uptake and partitioning ?

Publications

- DeRieux, W.-S. W., Li, Y., Lin, P., Laskin, J., Laskin, A., Bertram, A. K., Nizkorodov, S. A., and Shiraiwa, M.*: Predicting the glass transition temperature and viscosity of secondary organic material using molecular composition, ***Atmos. Chem. Phys. Discuss.***, https://doi.org/10.5194/acp-2017-1066, 2017.

- Mu. Q., M. Shiraiwa, M. Octaviani, N. Ma, A. Ding, H. Su, G. Lammel, U. Pöschl & Y. F. Cheng, Temperature effect on aerosol phase state and reactivity controls atmospheric multiphase chemistry and transport of polycyclic aromatic hydrocarbons, ***Science Advances***, in press, 2018.

- Hinks, M. L., Montoya-Aguilera, J., Ellison, L., Lin, P., Laskin, A., Laskin, J., Shiraiwa, M., Dabdub, D., and Nizkorodov, S. A.: Effect of Relative Humidity on the Composition of Secondary Organic Aerosol from Oxidation of Toluene, ***Atmos. Chem. Phys.***, 18, 1643-1652, 2018.