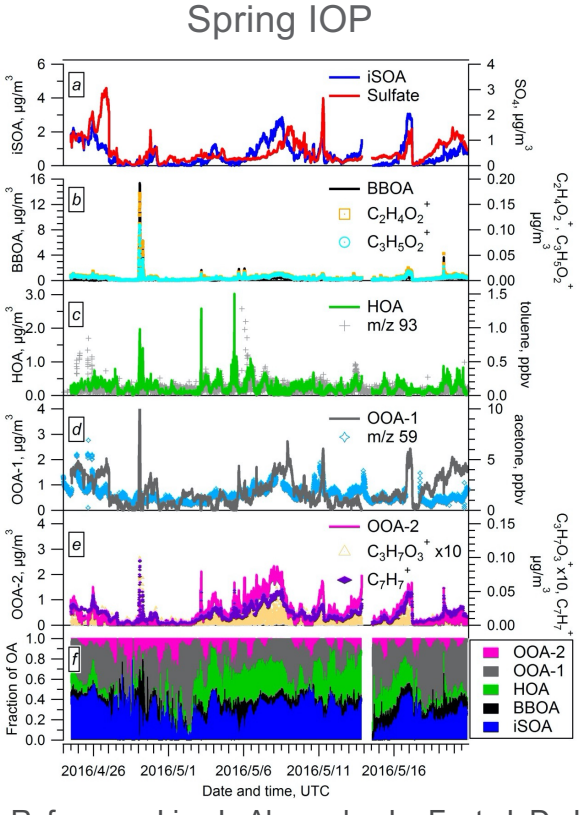
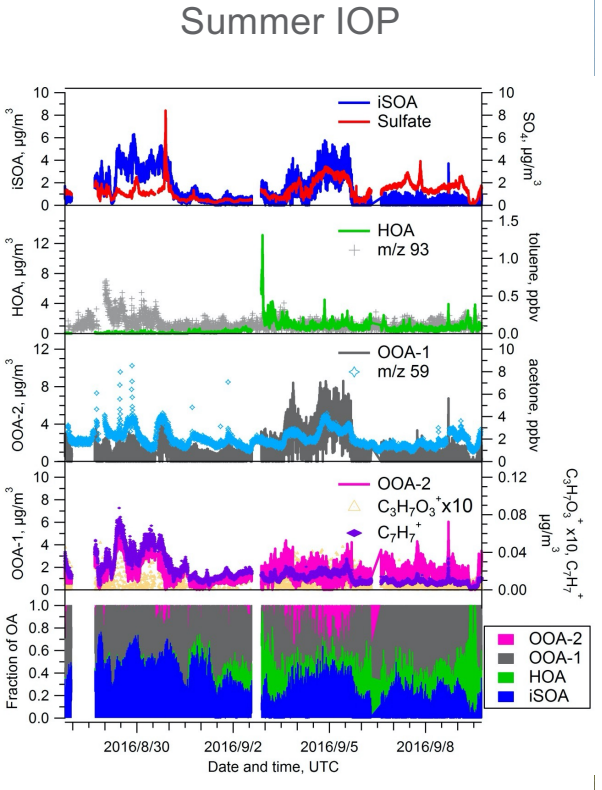
Organic Aerosol Characteristics at the SGP Site During HI-SCALE

- Aerosol loading higher in summer than spring.
- OA largest fraction of total.
- Quantified IEPOX and HOA at the SGP site for the first time.
- Most of the total OA is SOA.
- OA was more oxidized in the spring IOP (OSc = 0.29) than summer (OSc = -0.34).
- Unable to extract BBOA in summer.
- IEPOX SOA was more oxidized at SGP than other locations, likely due to weak local isoprene emissions.

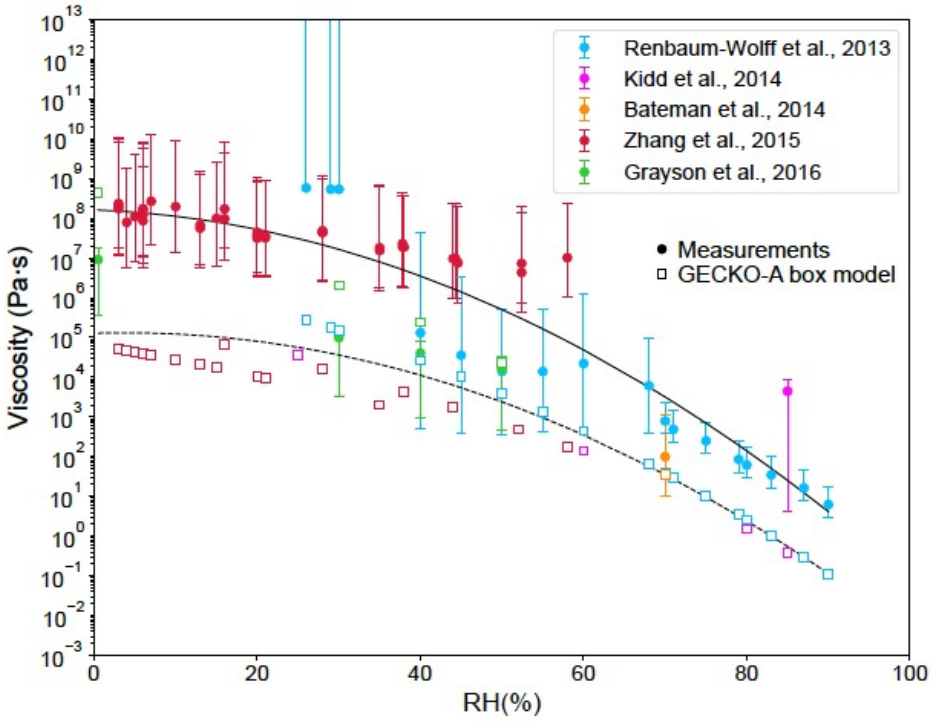




Reference: Liu, J., Alexander, L., Fast, J. D., Lindenmaier, R., and Shilling, J. E.: Aerosol characteristics at the Southern Great Plains site during the HI-SCALE campaign, Atmos. Chem. Phys., 21, 5101–5116, https://doi.org/10.5194/acp-21-5101-2021, 2021

Viscosity of SOA & Gas-Particle Partitioning

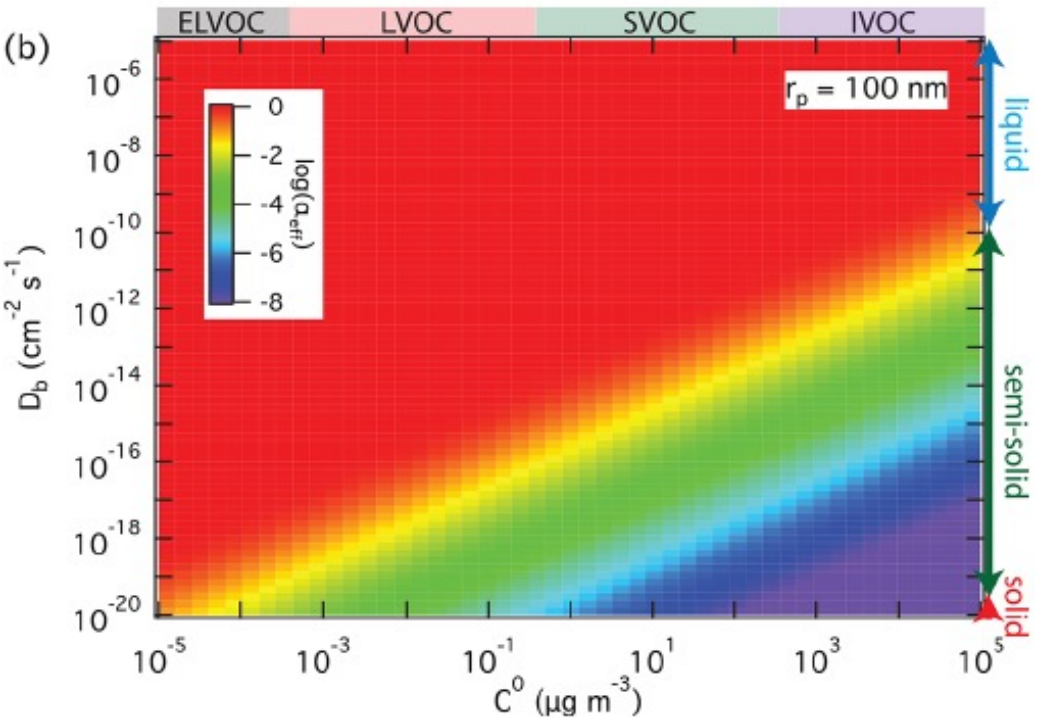
Phase State of SOA (liquid, semisolid, glassy)



SOA viscosity estimations by detailed gas-phase modeling (GECKO-A), reproducing viscosity measurements of α-pinene SOA

Galeazzo, T., Valorso, R., Li, Y., Camredon, M., Aumont, B. and Shiraiwa, M.: Estimation of Secondary Organic Aerosol Viscosity from Explicit Modeling of Gas-Phase Oxidation of Isoprene and α-pinene***, Atmos. Chem. Phys.***, https://doi.org/10.5194/acp-2021-5117, in press, 2021.

Effective Mass Accommodation Coefficient α_{eff}

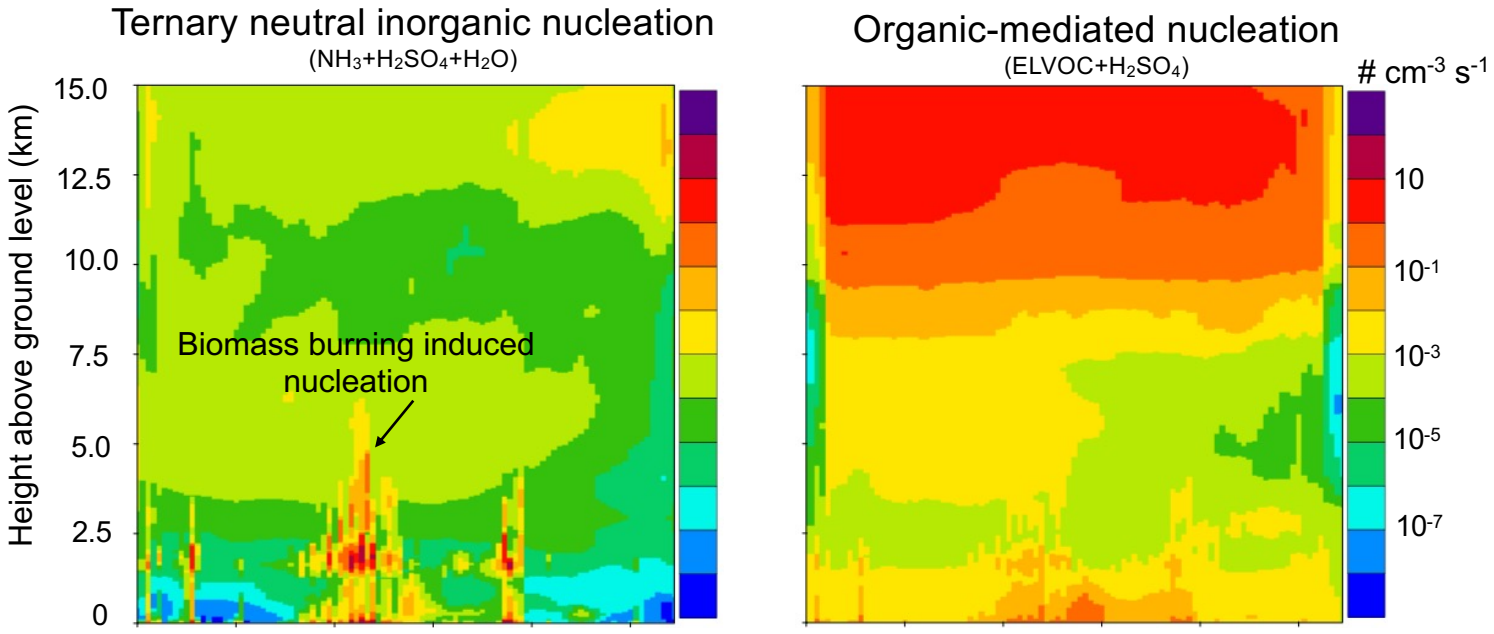


α_{eff} decreases substantially for semi-volatile compounds in semisolid particles

Kinetic limitations are likely important for glassy SOA in the free troposphere

Shiraiwa, M. and Pöschl, U.: Mass accommodation and gas–particle partitioning in secondary organic aerosols: dependence on diffusivity, volatility, particle-phase reactions, and penetration depth, ***Atmos. Chem. Phys.***, 21, 1565-1580, 2021.

WRF-Chem: Nucleation rates in biomass burning plumes



- Biomass burning induced nucleation extends from surface to 5 km altitude due to wildfire plume rise and is mainly ternary-inorganic-neutral
- SOA dominates ultrafine particle mass, 20% is BB-SOA rest is biogenic SOA
- Primary BBOA is 40% of accumulation particle mass, but less than 2% of ultrafine mass

Transport and chemistry of isoprene in deep convective clouds in an LES Framework

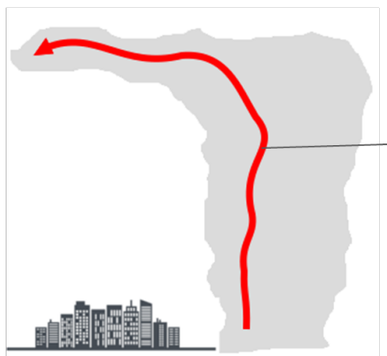
R. Bardakov, **J. A. Thornton**, I. Riipinen, R. Krejci, A. M. L. Ekman

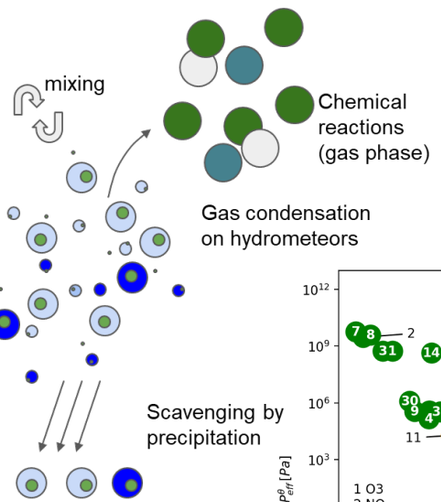
UNIVERSITY of WASHINGTON

Stockholm University

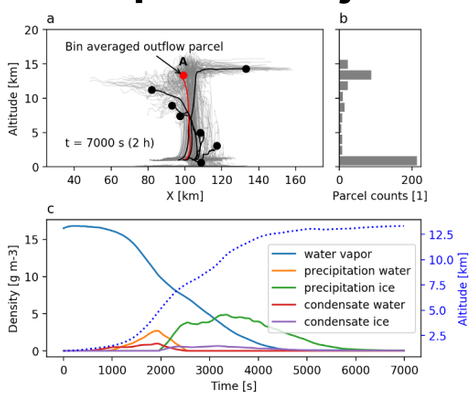
In review (Tellus)

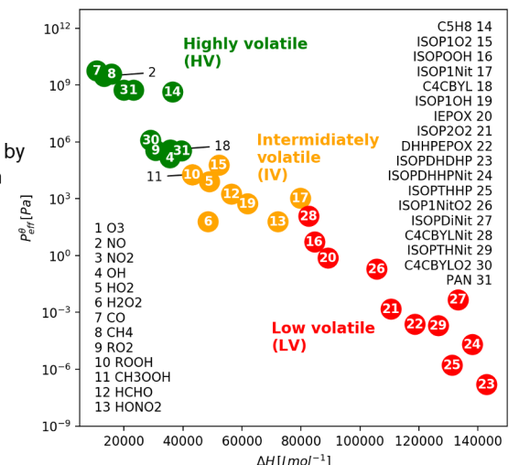
Trajectories framework





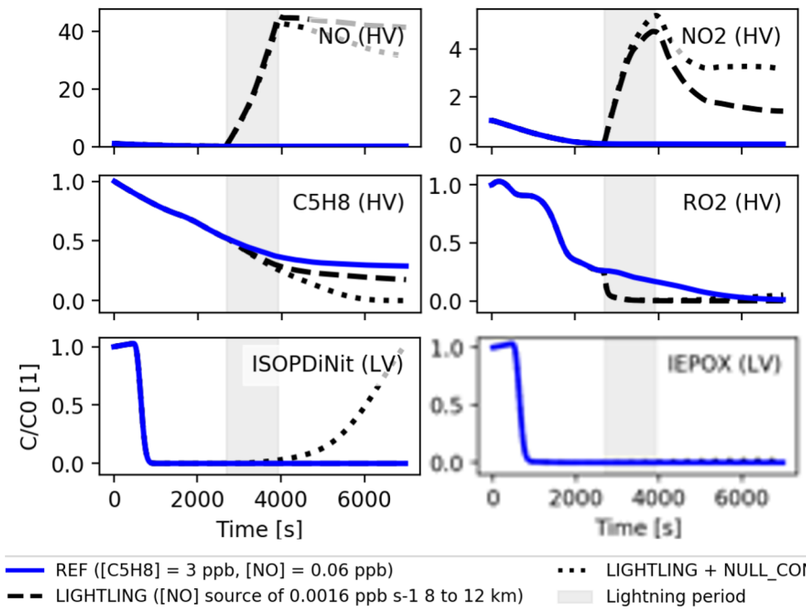
400 air parcel trajectories





31 oxidants, isoprene and oxidation products spanning wide volatility range

4 hydrometeor types



- lifetime of isoprene in outflow and volatility distribution determined by lightning NO_x
- condensation of low volatility products to anvil ice is major uncertainty for new particle growth
- simulations with detailed monoterpene & isoprene mechanisms now in progress

Mapping chemical complexity in lab and field OA using UCB GLOBES mass spectral library

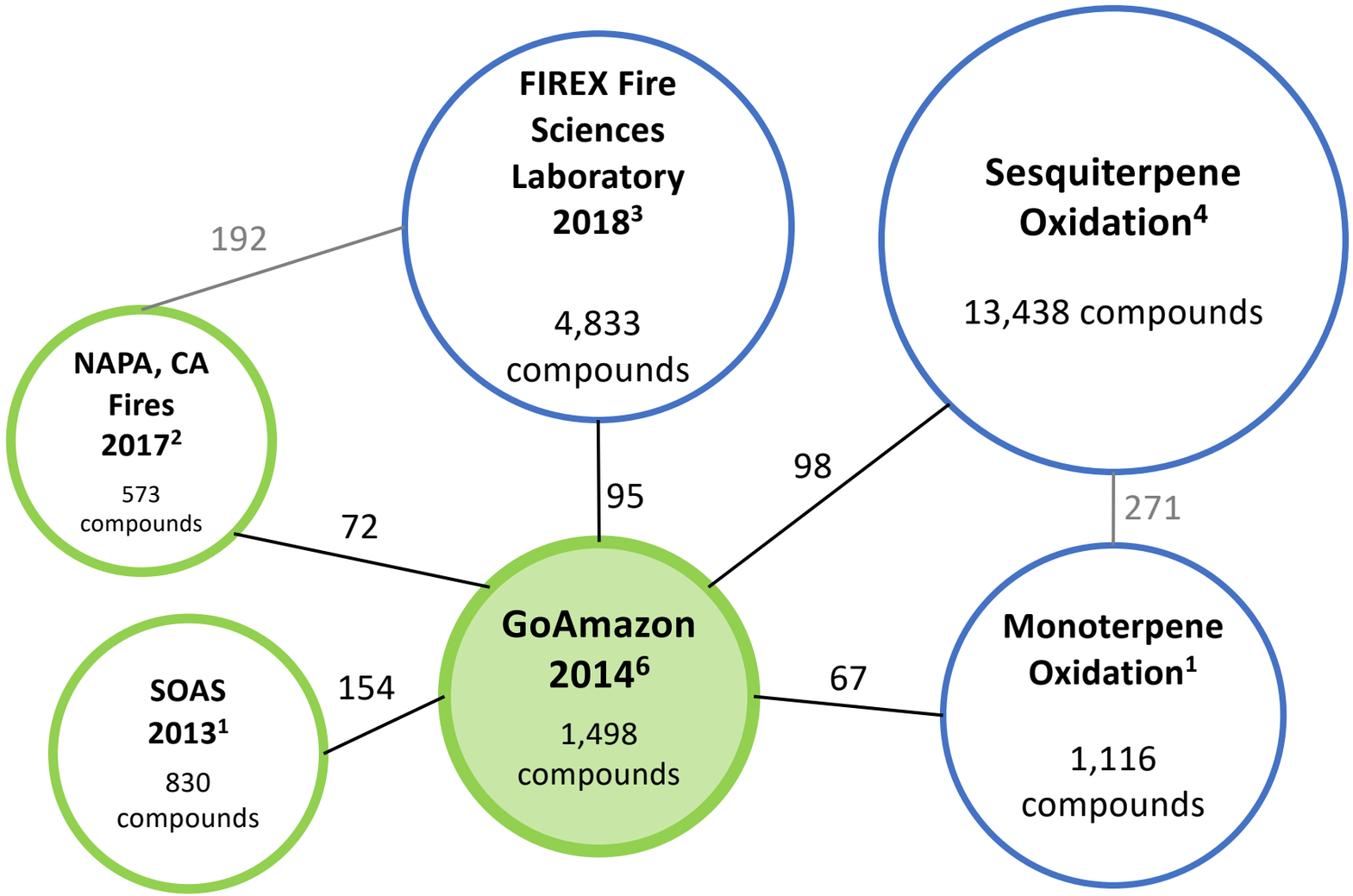
Lindsay Yee, Emily Barnes, Robin Weber, and Allen Goldstein

Preliminary analyses:

1) Curating ~24,000 mass spectra; which are novel tracers?
2) More chemical diversity in lab-generated vs field-observed OA
3) Majority of observed compounds (e.g. 77% in GoAmazon) still unique and unidentified

UCB GLOBES EI mass spectral library (open-access):

https://nature.berkeley.edu/ahg/resources/



[1]Zhang et al., PNAS, 2018; [2]Liang et al., ACP, 2021; [3]Jen et al., ACP, 2019; [4]Yee et al., ACP, 2018; [6]Barnes et al., in prep.

Effect of Particle Phase State on SOA Partitioning as a Function of Relative Humidity

Objective

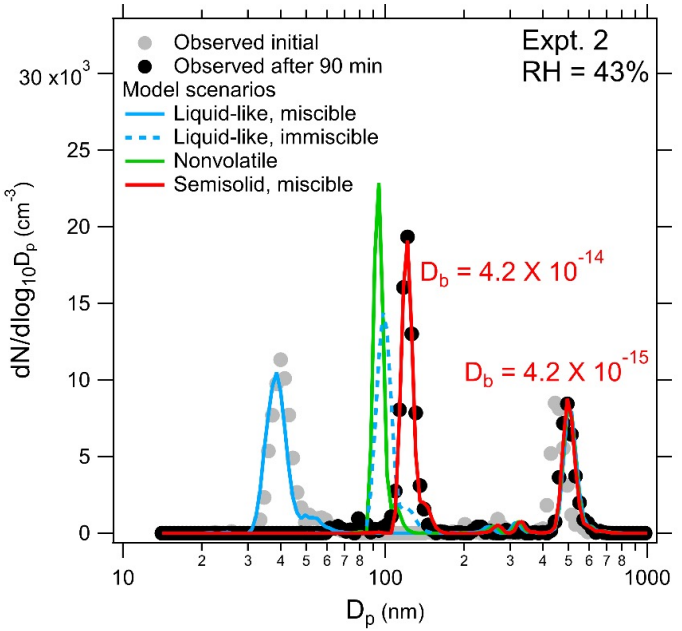
- Investigate the role of phase state and bulk diffusivity in partitioning of semivolatile organic compounds to aged secondary organic aerosol

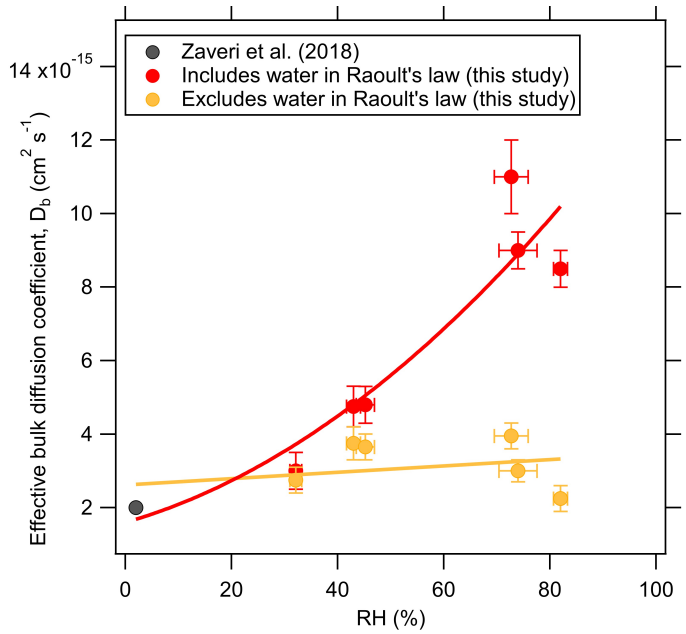
Approach

- Observed growth kinetics of secondary organic aerosol formation in an environmental chamber at different RH from photooxidation of isoprene in the presence of preexisting Aitken mode (potassium sulfate) and accumulation mode (aged α-pinene SOA)
- Interpreted the data with the MOSAIC aerosol model to unravel the effects of bulk diffusivity on the size-dependent growth kinetics of secondary organic aerosol formation

Finding

- Data analyses show isoprene semivolatile organic chemicals favor growth of small particles due to low diffusivity inside larger, aged secondary organic aerosols up to 80% RH.





Zaveri RA, JE Shilling, A Zelenyuk, MA Zawadowicz, K Suski, S China, DM Bell, D. Veghte, and A Laskin, Particle-phase diffusion modulates partitioning of semivolatile organic compounds to aged secondary organic aerosol, *Environ. Sci. & Technol.*, 54: 2595-2605, 2020.

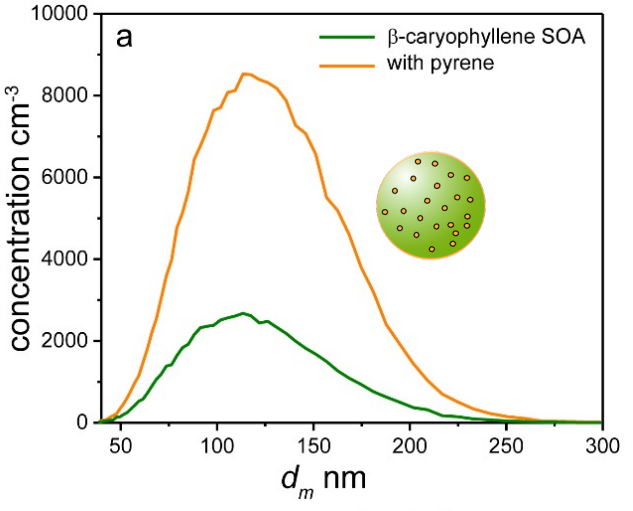
Synergetic Interactions between PAHs and Biogenic SOA Particles

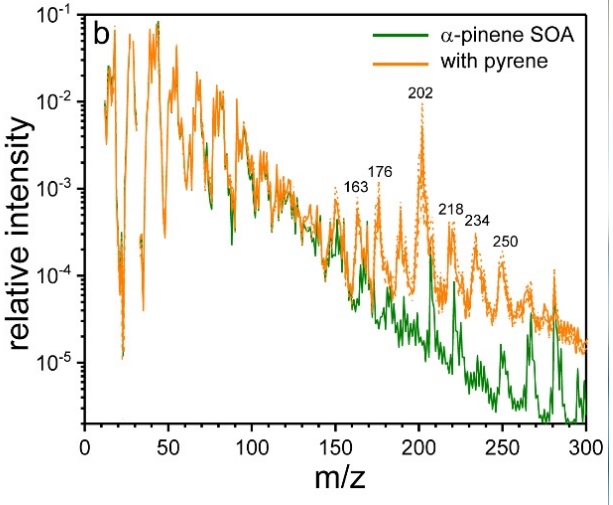
- The presence of PAHs during BSOA formation yields significantly higher particle number and mass concentrations
- PAH-SOA particles contain trapped, unreacted PAHs, and products of heterogeneous reactions between PAHs with ozone, enabling their LRT
- SOA particles formed in the presence of PAHs (PAH-SOA) are more viscous, less volatile, **contains more oligomers**

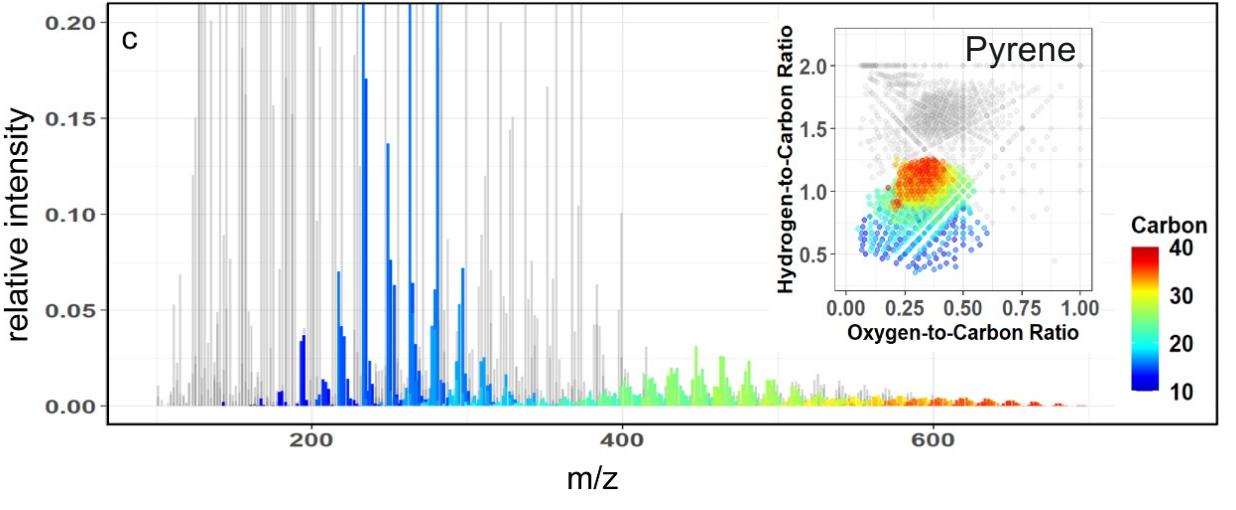
- The "extra mass" is dominated by oxidation products of biogenic VOCs
- Observed two distinct pathways for PAH-enhanced oligomer formation

Pyrene: C_{16}-C_{26}-C_{36}

Benzo(a)anthracene: C_{10}-C_{20}-C_{30}-C_{40}



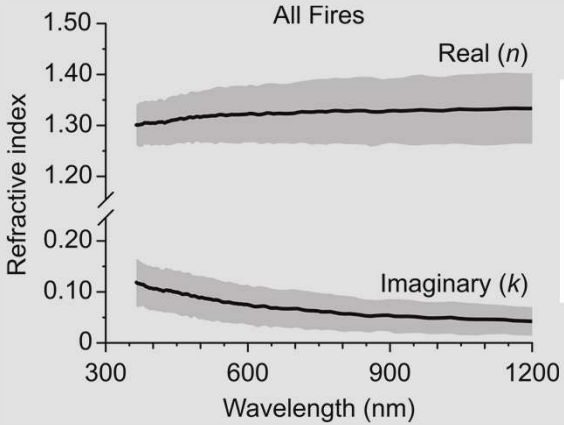


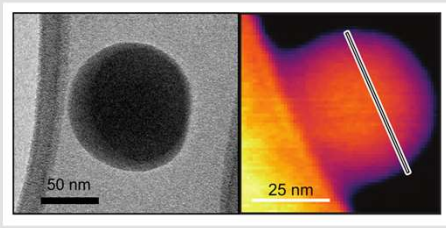


Implementation of FIREX-AQ measured optical properties in WRF-Chem for estimations of secondary organic aerosol (SOA) formation

Chenchong Zhang, Nishit Shetty, Benjamin J. Sumlin, Rajan K. Chakrabarty*

Washington University in St. Louis





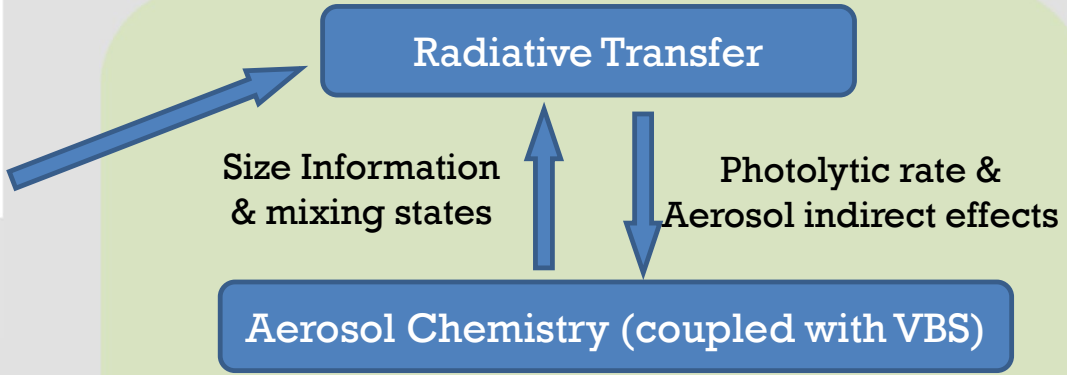
OA refractive indices and microstructure detected by STEM-EELS

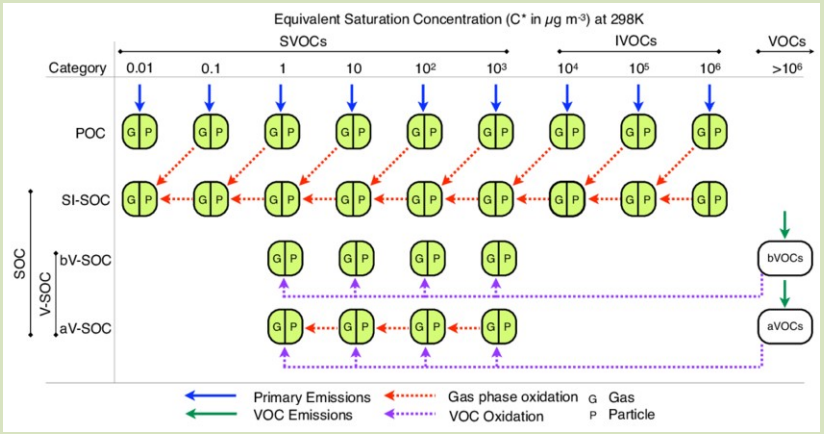
Main results from FIREX-AQ campaign (Jul-Sep, 2019):

- The refractive indices measured by electron energy-loss spectroscopy (EELS) and PAS show higher light absorption of OA in shortwave wavelengths than previously reported values.

Expected outcomes:

1. Insights into aerosol-meteorology feedback mechanism.
2. Evaluation of aerosol radiative effects and SOA formation based on observational dataset.





Schematic for the VBS framework (Jathar, et al., ACP, 2011)

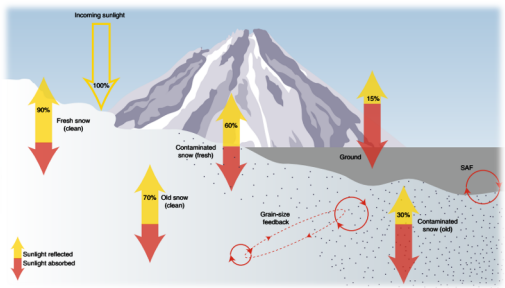
Radiative Effects of Organic Carbon Deposited on Snowpack

- *Yue Zhou, Alexander Laskin, et al. (ARM/ASR PI meeting, June 23, 2021)*

PURDUE UNIVERSITY

Motivation

- Composition-specific influence of OC on the snow albedo reduction.

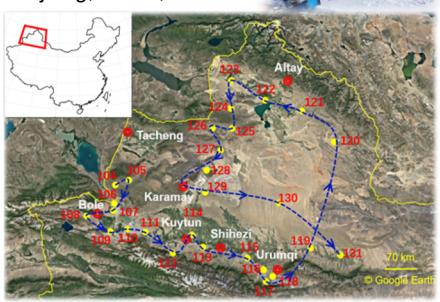


https://doi.org/10.1038/s41558-018-0296-5

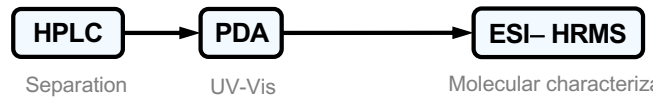
Snow sampling

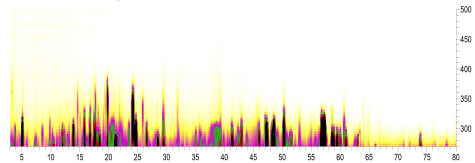
蘭州大學 Lanzhou University

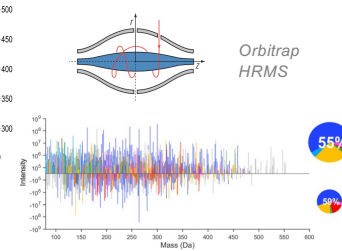
- Xinjiang, China; 28 sites



Composition-specific optical properties of snow BrC

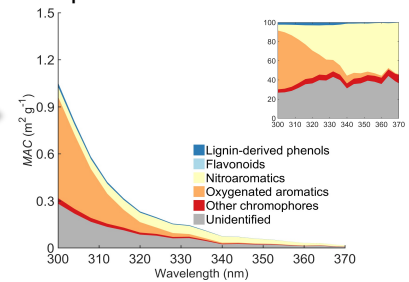






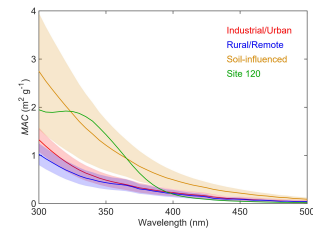
- separation and detection of strong chromophores.

- relative contributions of different types of chromophores to the total *MAC*.

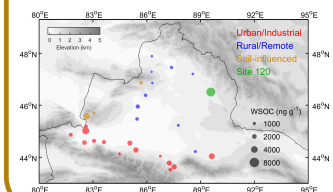


Radiative forcing of snow BrC

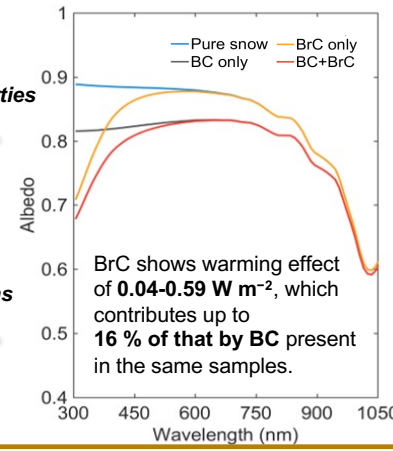
https://doi.org/10.5194/acp-21-8531-2021



BrC optical properties



WSOC concentrations



BrC shows warming effect of **0.04-0.59 W m^{-2}**, which contributes up to **16 % of that by BC** present in the same samples.

Snow physical properties

Model simulation

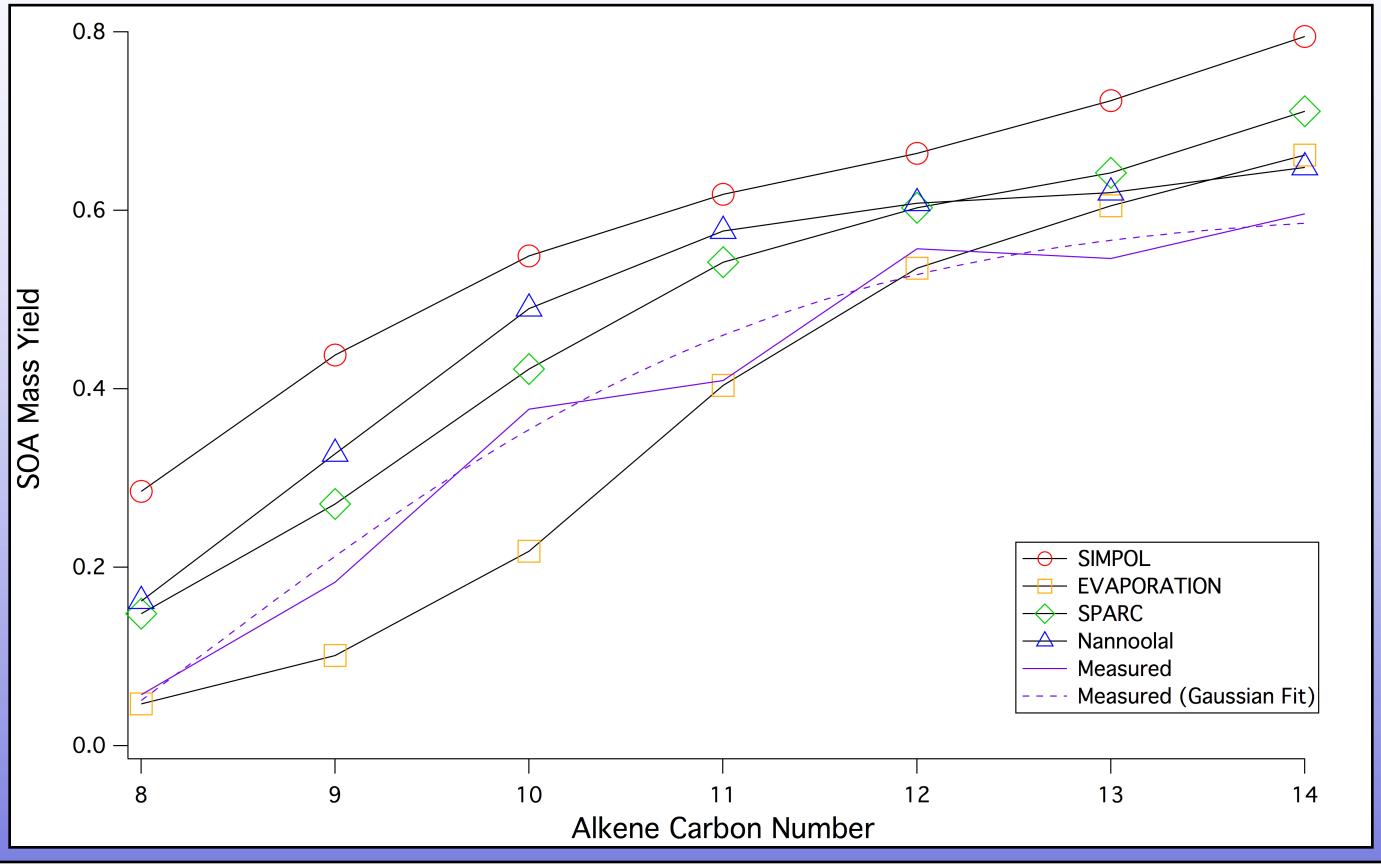
EarthSciCode/ **SNICARv2**

SNICAR snow model codes (for educational & research purposes only)

Snow, Ice, and Aerosol Radiation (SNICAR) model

Evaluation of Four Vapor Pressure Estimation Methods Using a Highly Constrained Model for SOA Formation from Reactions of Alkenes + OH/NO_x

Emmaline Longnecker, Julia Bakker-Arkema, Jose-Luis Jimenez, Paul Ziemann
Department of Chemistry and CIRES, University of Colorado Boulder



ASR Atmospheric System Research

University of Colorado Boulder

CIRES

- Explicit, quantitative, gas-phase mechanism for C_8–C_{14} 1-alkenes + OH/NO_x
- Products: dihydroxycarbonyls, dihydroxynitrates, hydroxynitrates, hydroxycarbonyls, alkyl nitrates, & aldehydes – no oligomers
- Gas-wall & gas-particle partitioning & secondary OH reactions