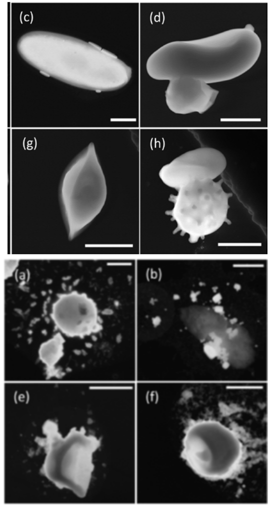
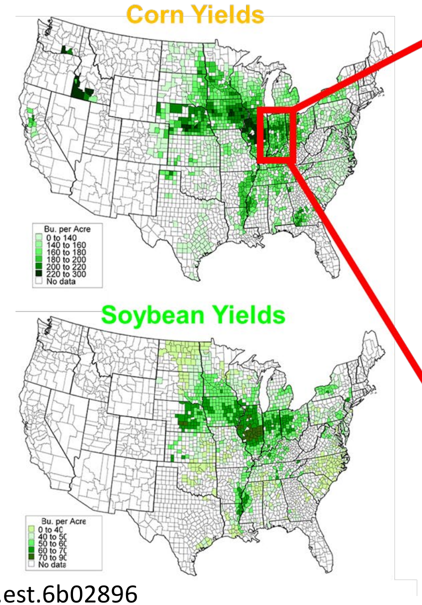
Chemical Imaging of Vertically Resolved Atmospheric Particles Collected in Past and Ongoing Field Studies

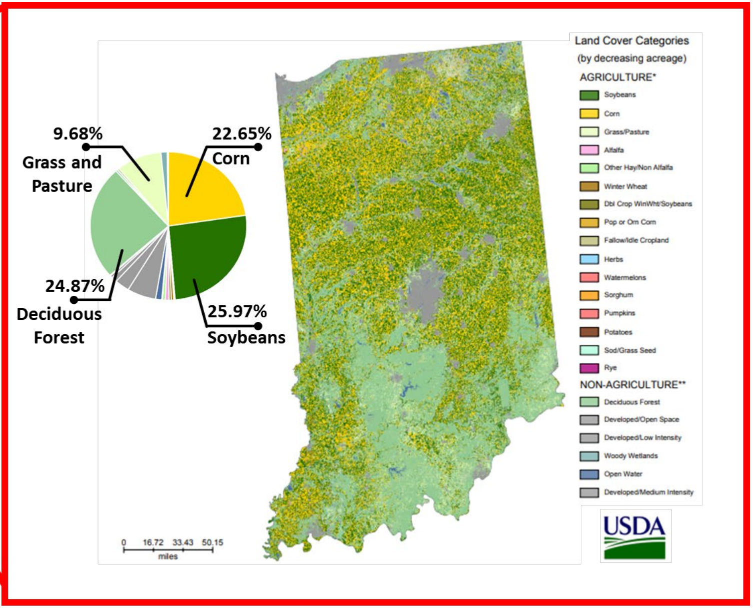
A. Laskin, *Purdue University*

Airborne Biological Particles from Crops Harvesting

- emissions <u>vary with plant life cycle</u>
- break down during RH cycling, <u>release accumulation mode fragments</u>







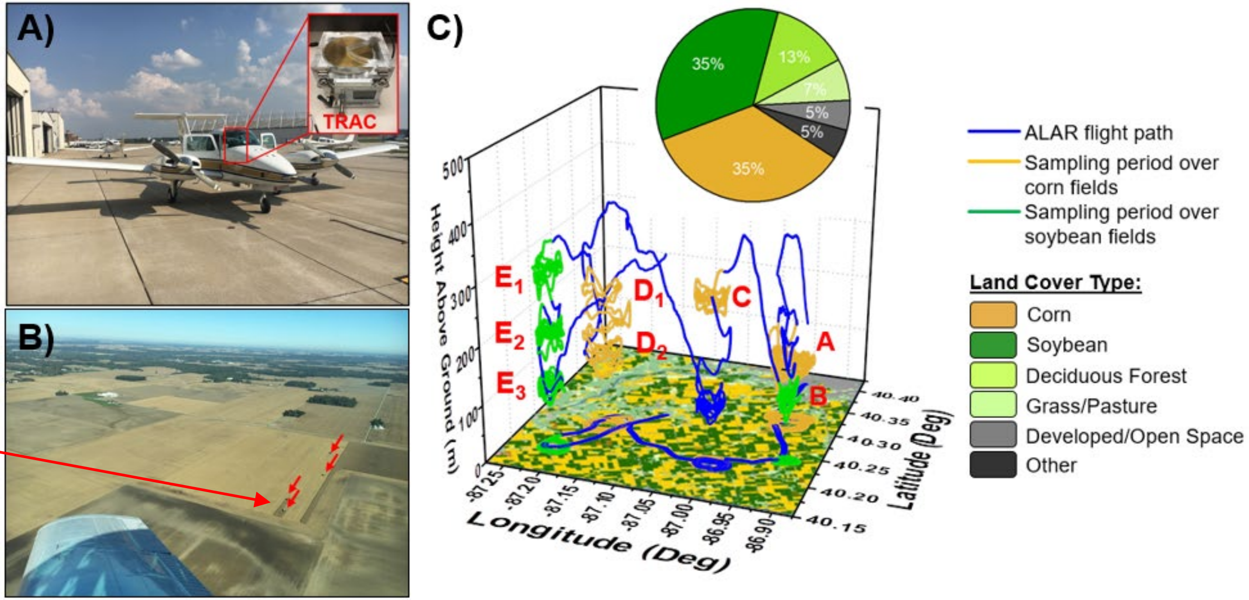
China et al, 2016, https://pubs.acs.org/doi/10.1021/acs.est.6b02896

PURDUE UNIVERSITY

Tomlin et al, 2020, https://dx.doi.org/10.1021/acsearthspacechem.0c00172

Airborne Biological Particles from Crops Harvesting

- emissions vary with plant life cycle
- break down during RH cycling, release accumulation mode fragments

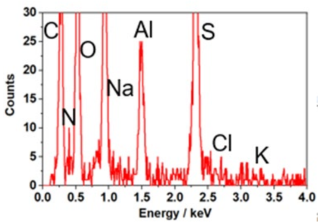


Purdue University Airborne Laboratory for Atmospheric Research (ALAR)
P. Shepson, PI

PURDUE UNIVERSITY

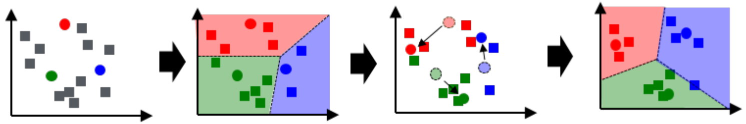
Tomlin et al, 2020, https://dx.doi.org/10.1021/acsearthspacechem.0c00172

Particle-Type Composition

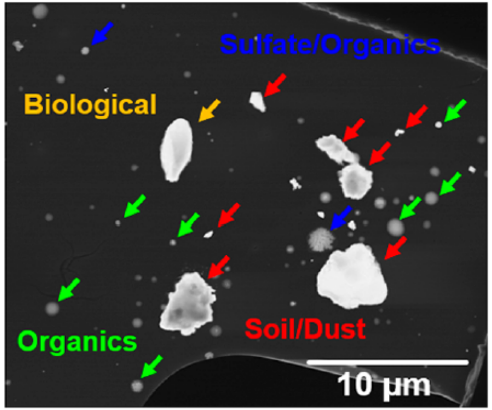


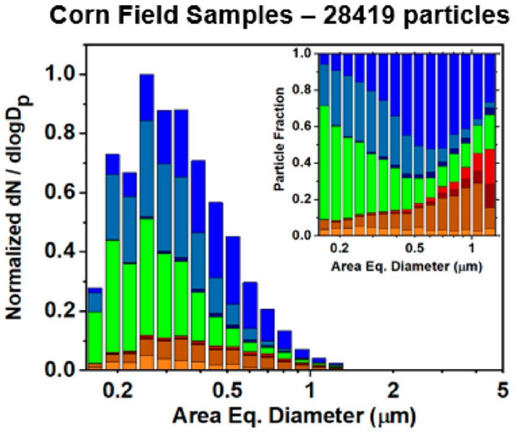
EDX spectra

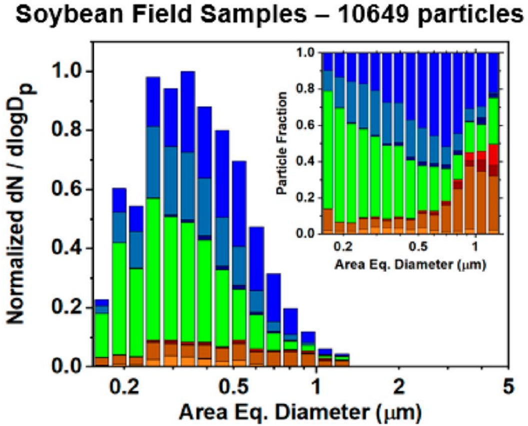
Unsupervised machine learning algorithm: K-means clustering



CO organics CNO/S CO/S Ca-dust AlSi-dust Biological I, II







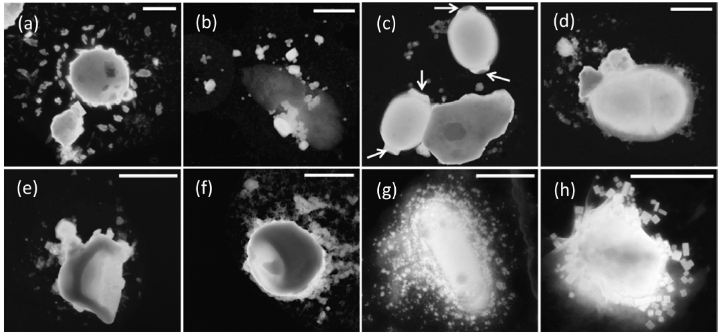
- **Biological particles are 40-50% of >1.0 µm particles**
  5-10% of <0.5 µm particles

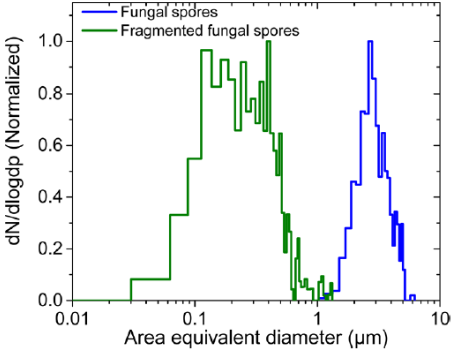
PURDUE UNIVERSITY

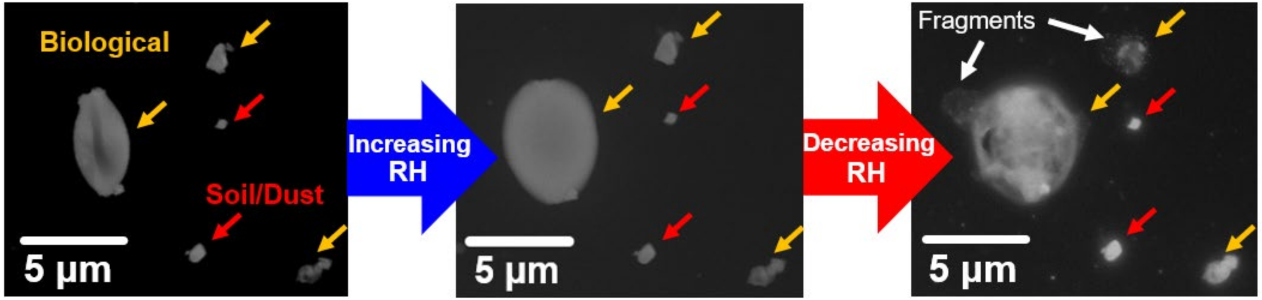
Tomlin et al, 2020, https://dx.doi.org/10.1021/acsearthspacechem.0c00172

Disintegration of Biological Particles

China et al, 2016, https://pubs.acs.org/doi/10.1021/acs.est.6b02896







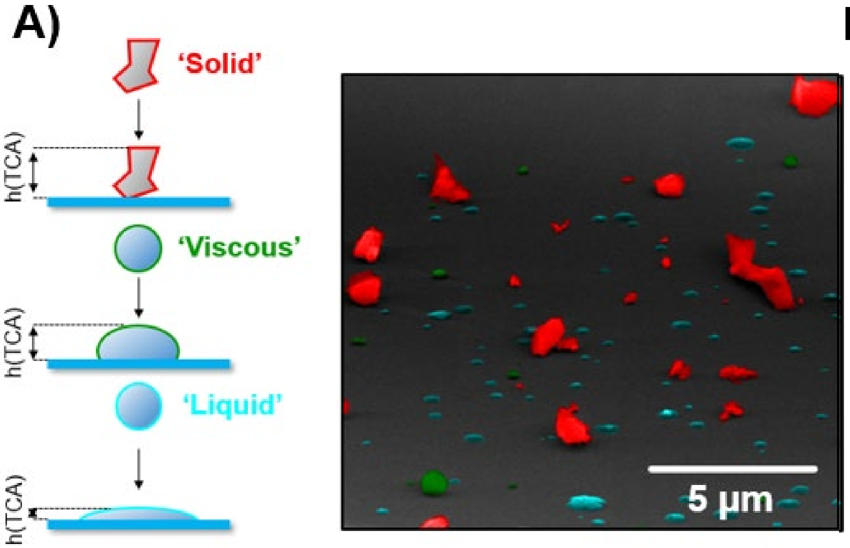
Environmental SEM experiment imaging particles in a hydration/dehydration cycle.
Biological particles disintegrate and release smaller particles

PURDUE UNIVERSITY

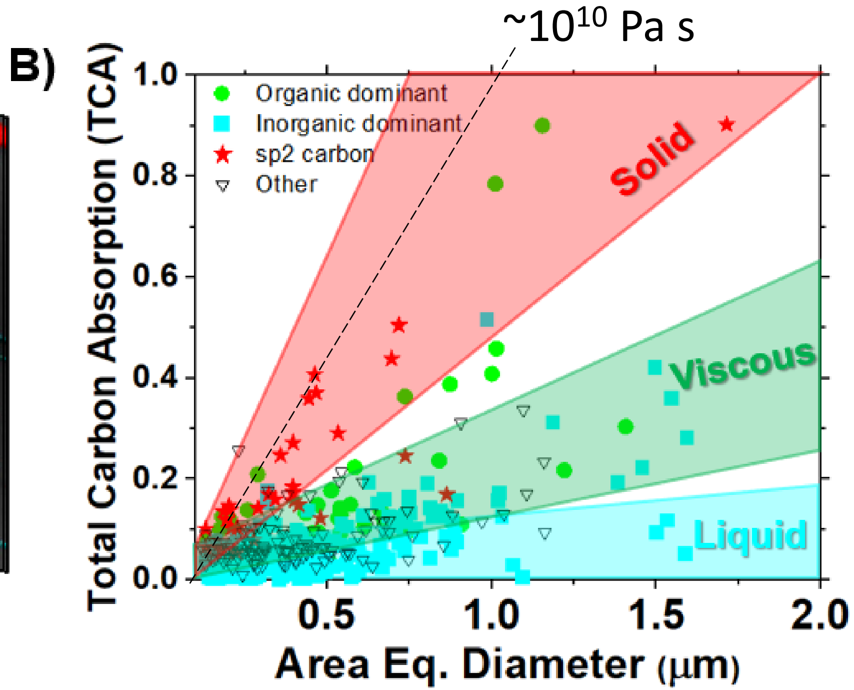
Tomlin et al, 2020, https://dx.doi.org/10.1021/acsearthspacechem.0c00172

Solid Phase of Biological Particles

- **Biological particles are solid, plausibly IN active**



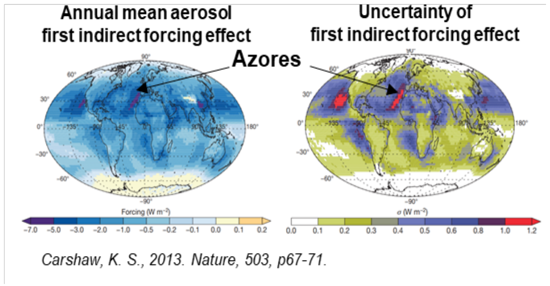
75° tilted SEM image

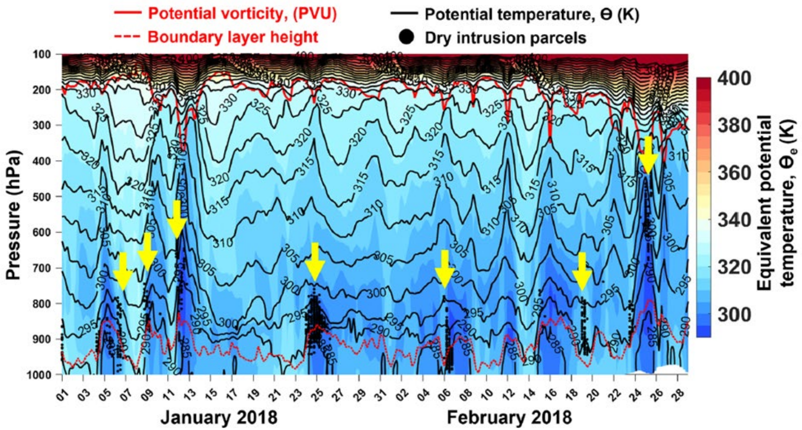


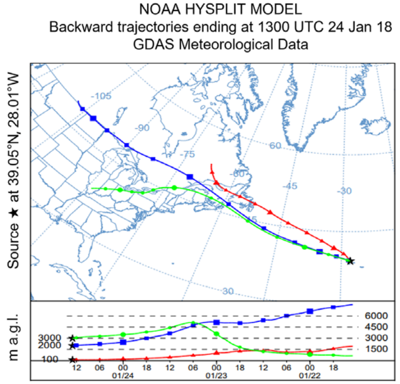
Particle height is related to total carbon absorption (TCA) obtained from NEXAFS spectra

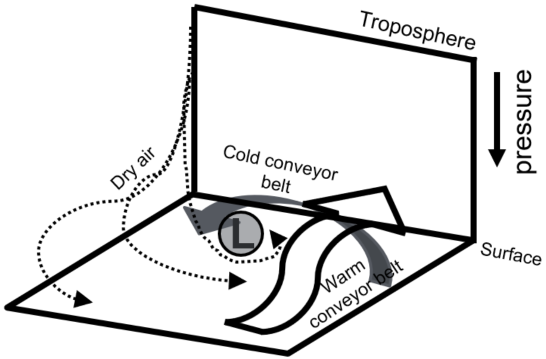
Tomlin et al, 2020, https://dx.doi.org/10.1021/acsearthspacechem.0c00172

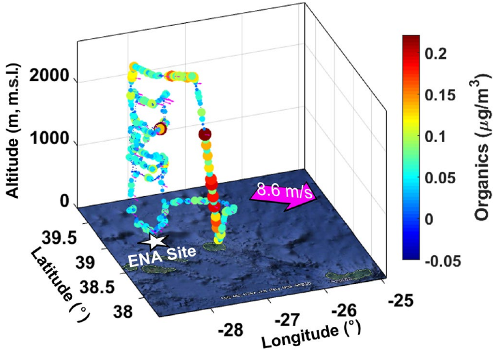
ACE-ENA 2018 - Dry Intrusions Events











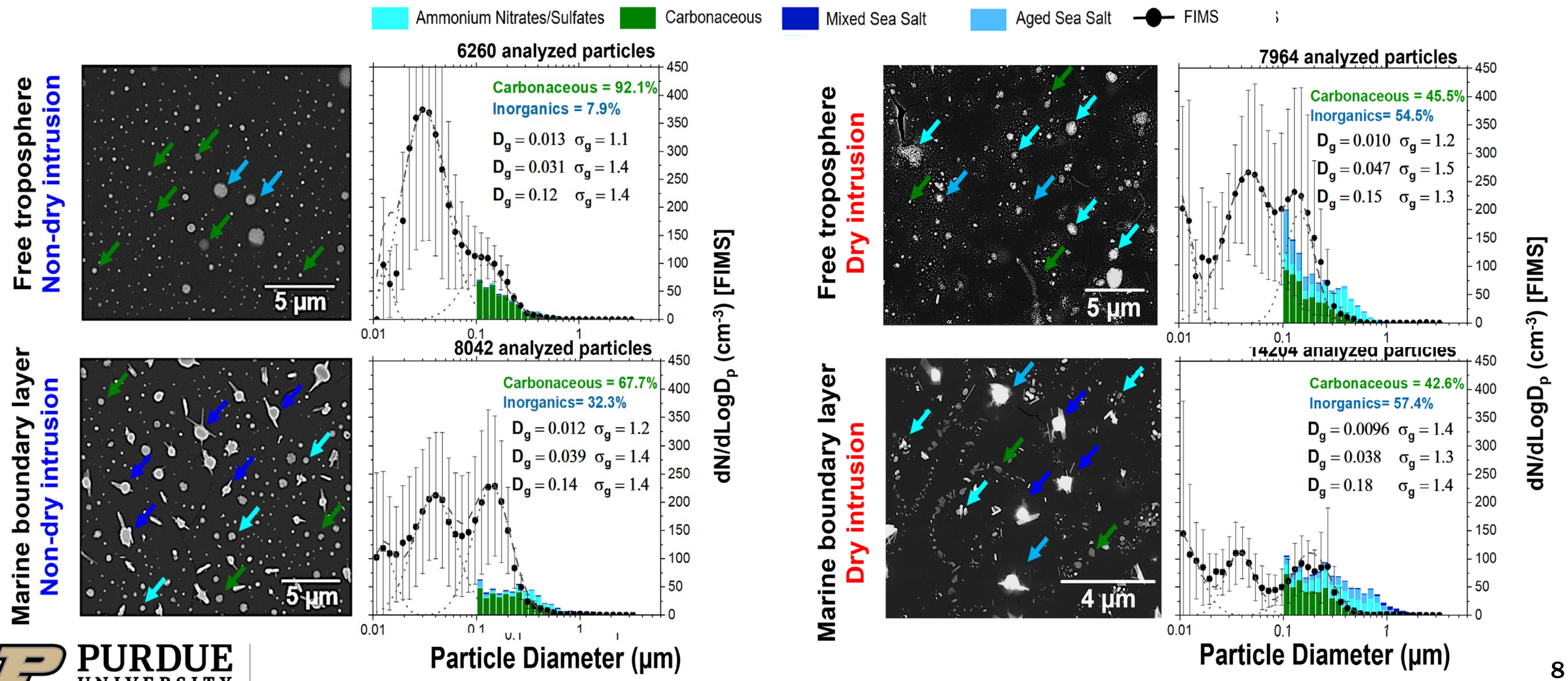
PURDUE UNIVERSITY

Tomlin et al, 2021, https://doi.org/10.5194/acp-21-18123-2021

Particle-Type Composition from CCSEM/EDX

- 6 samples from non-dry intrusion FT
- 10 samples from non-dry intrusion MBL

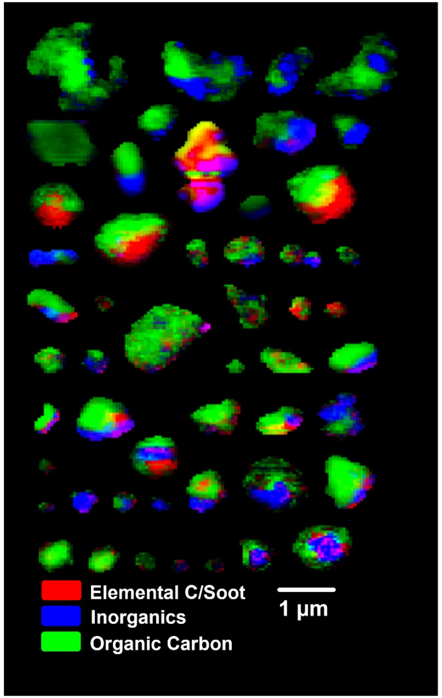
- 8 samples from dry intrusion FT
- 14 samples from dry intrusion MBL

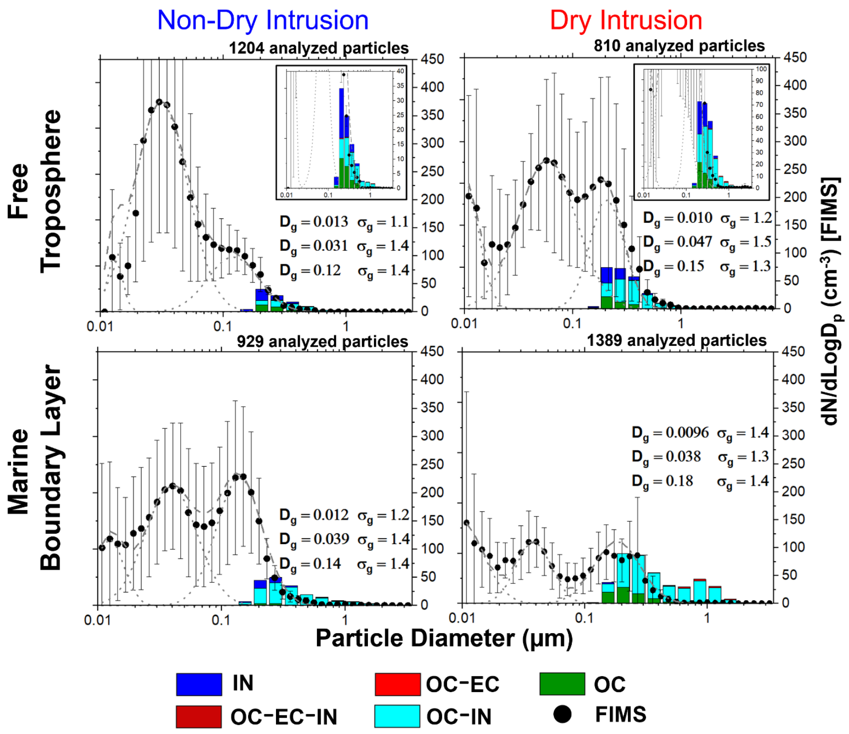


PURDUE UNIVERSITY

Tomlin et al, 2021, https://doi.org/10.5194/acp-21-18123-2021

Particle Internal Mixing State from STXM/NEXAFS





- **DI particles are more diverse, both externally and internally**

Tomlin et al, 2021, https://doi.org/10.5194/acp-21-18123-2021

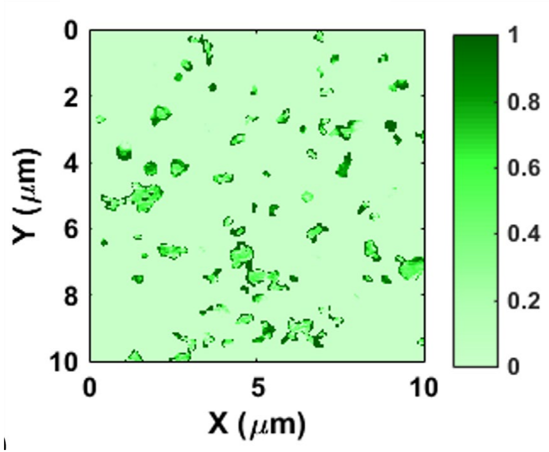
Particle Organic Volume Fractions → κ values

Assessment of Particles Hygroscopicity (κ)

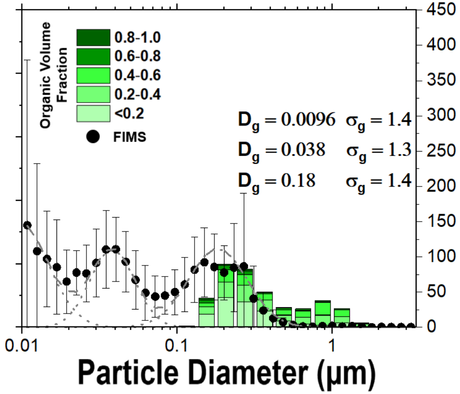
$$\kappa = (1 - OVF_{org})\,\kappa_{inorg} + OVF_{org}\,\kappa_{org}$$

0.6 for $(NH_4)_2SO_4$

0-0.1 for organics



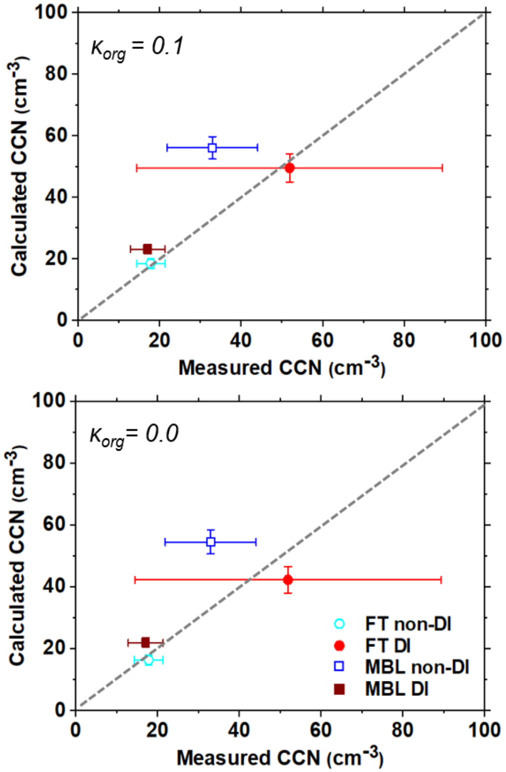
Fraund et. al. AMT. 12,1619. 2019.



$$\kappa_{FT,\ non\text{-}DI} = 0.36 \qquad \kappa_{MBL,\ non\text{-}DI} = 0.48$$

$$\kappa_{FT,\ DI} = 0.33 \qquad \kappa_{MBL,\ DI} = 0.41$$

κ – Calculated vs Measured



PURDUE UNIVERSITY

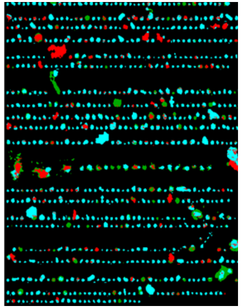
Tomlin et al, 2021, https://doi.org/10.5194/acp-21-18123-2021

Particle Mixing State Parameterization

→ **Parameterization using Shannon entropy metrics**

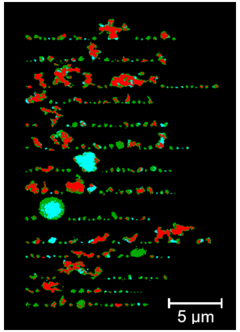
Riemer & West, 2013, https://doi.org/10.5194/acp-13-11423-2013

1



.
.
.
.

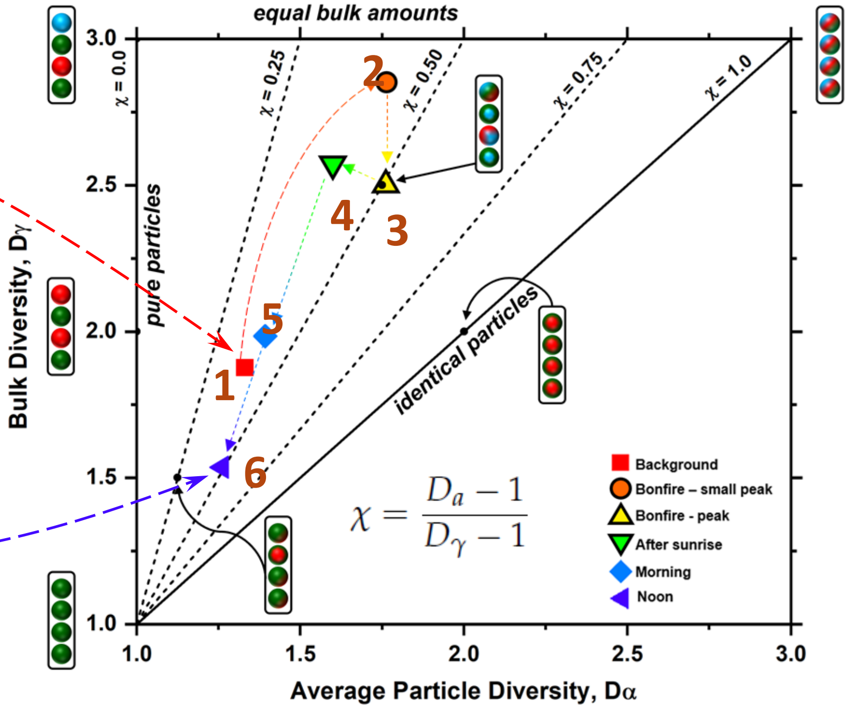
6



$D_i = e^{H_i}$ Number of species within a specific particle

$D_\alpha = e^{H_\alpha}$ Average number of species within any given particle

$D_\gamma = e^{H_\gamma}$ Number of species within the entire sample

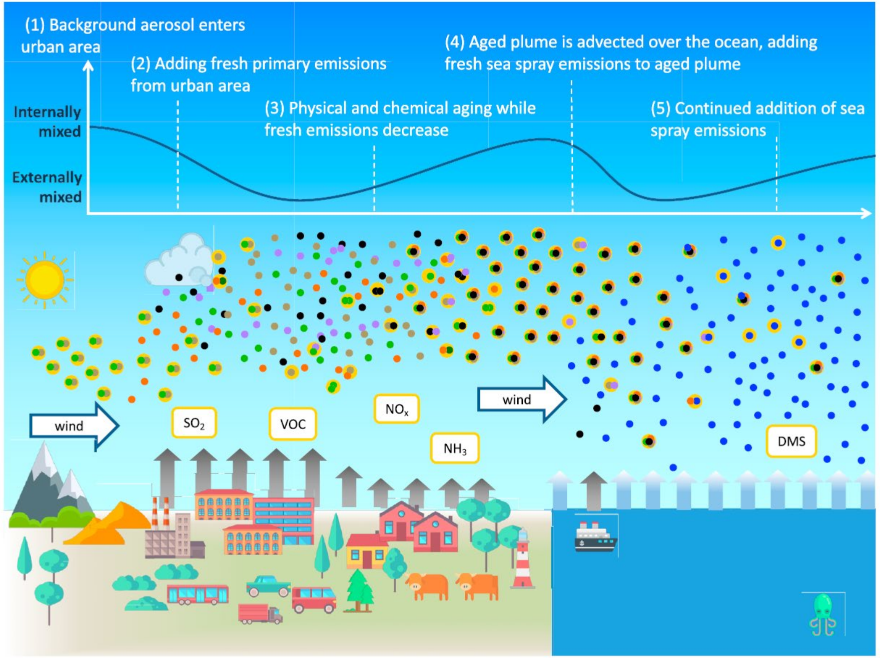


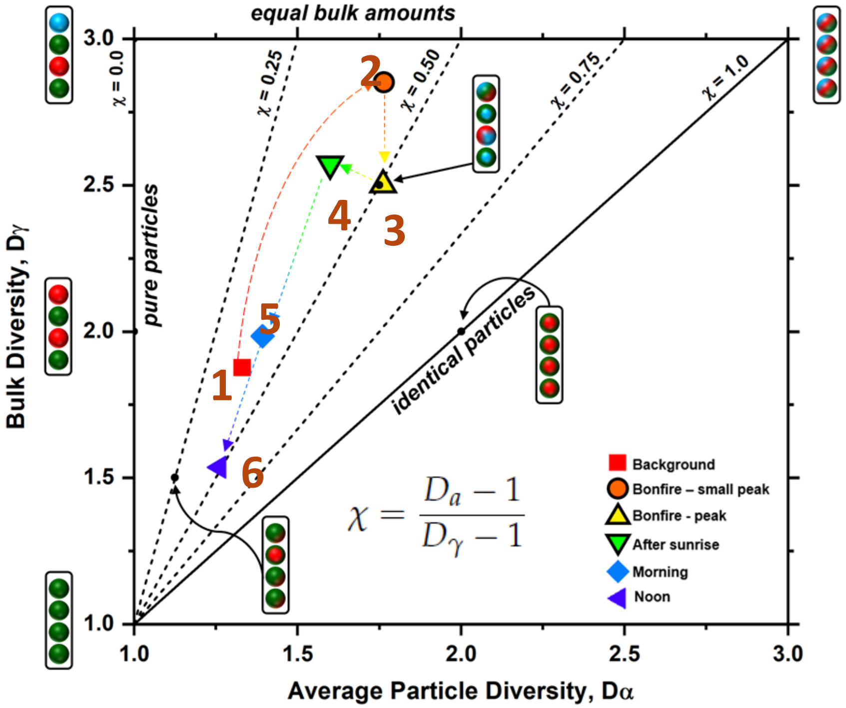
Tomlin et al, 2022,
https://pubs.rsc.org/en/content/articlepdf/2022/ea/d2ea00037g

Particle Mixing State Parameterization

Inform Particle-resolved Models ⟷ Parameterization using Shannon entropy metrics

Riemer et al, 2019, https://doi.org/10.1029/2018RG000615 **Riemer & West, 2013,** https://doi.org/10.5194/acp-13-11423-2013





Tomlin et al, 2022,
https://pubs.rsc.org/en/content/articlepdf/2022/ea/d2ea00037g

Ongoing Studies and Plans: SAIL

- **Parties from Tethered Balloon Sampling**

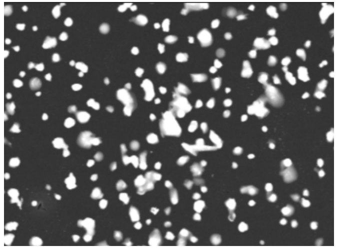
May 2022 flights

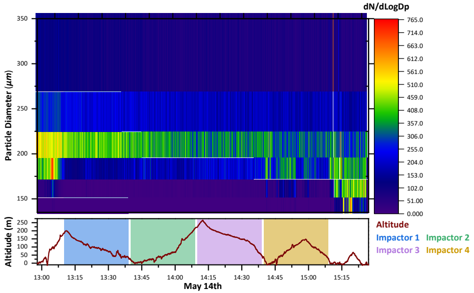
– Size and Time-resolved Aerosol Collector (STAC)

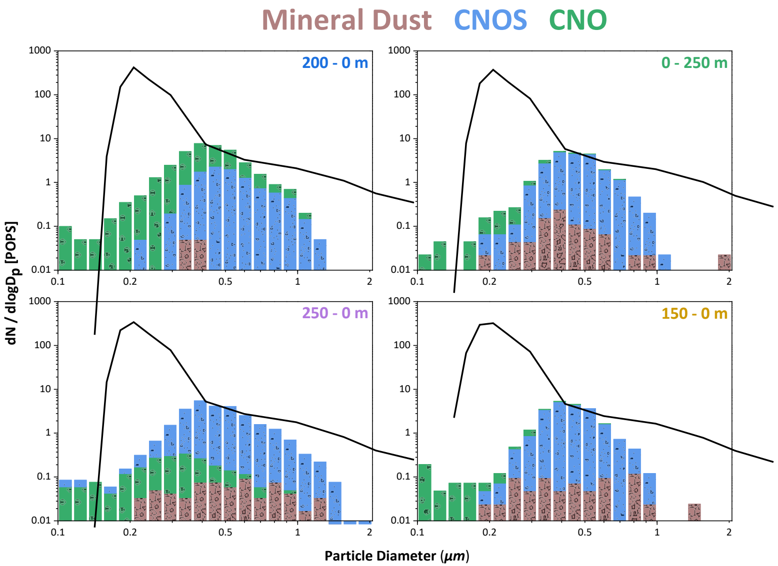
– T, P, RH, sensors

– OPC, particle size distribution

– a micro-aethalometer, BC mass loading



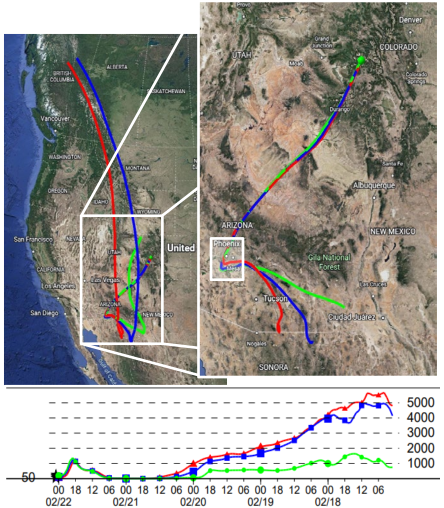




PURDUE UNIVERSITY | Polytechnic Institute

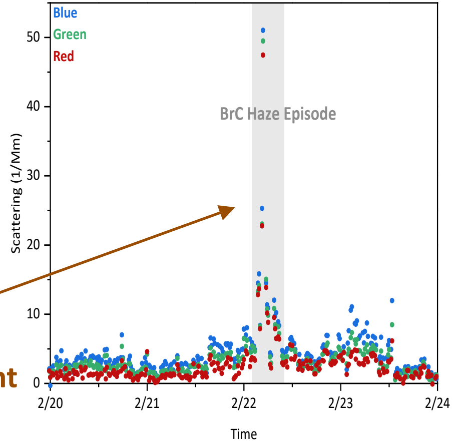
Ongoing Studies and Plans: SAIL

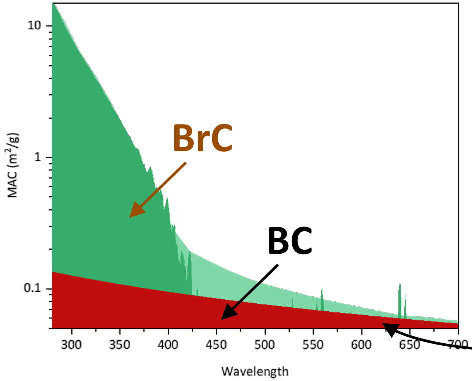
- **PM Deposits in Snow**

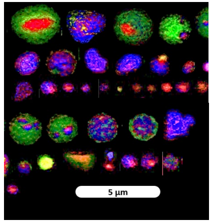


– **February 22, 2022**
descending trajectory from Western Rockies to Phoenix, low altitude transport to SAIL

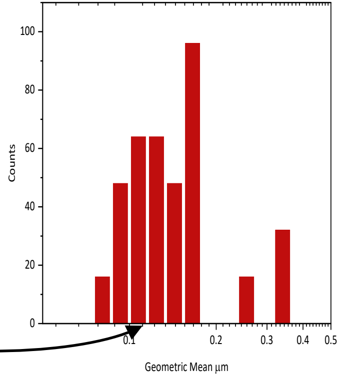
BrC
Haze Event







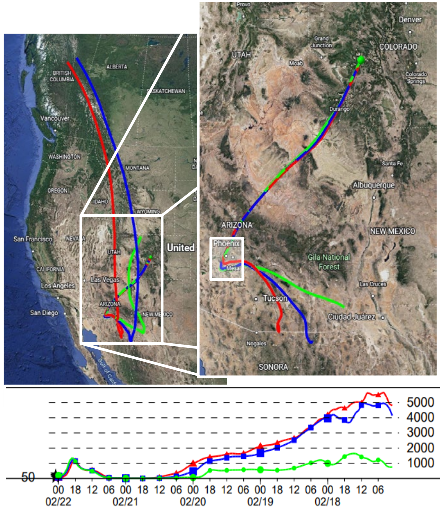
Insoluble BC deposits



PURDUE UNIVERSITY

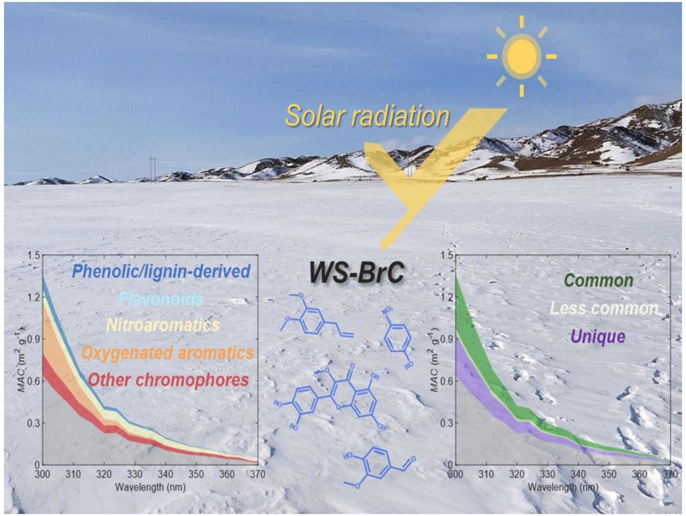
Ongoing Studies and Plans: SAIL

- **PM Deposits in Snow**

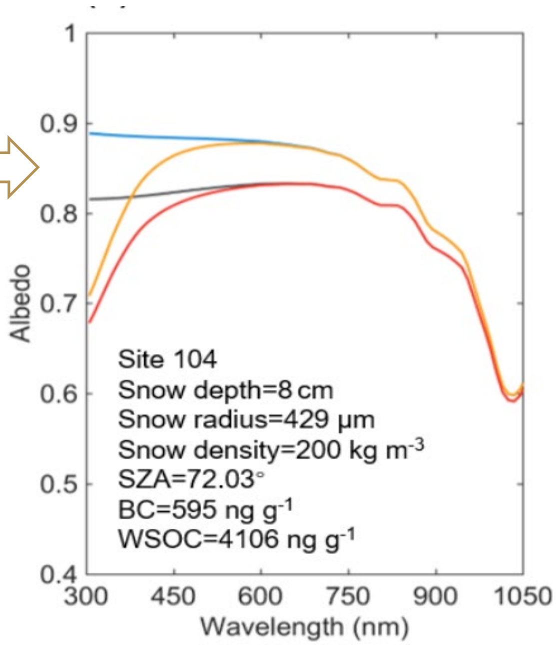


- **February 22, 2022**
descending trajectory from Western Rockies to Phoenix, low altitude transport to SAIL

Planned: Molecular Characterization of BrC
→ assessment of snow reflectivity



Zhou et al EST 2022, 56, 4173-4186



Zhou et al ACP 2021, 21, 8531-8555

PURDUE UNIVERSITY

References

J.M. Tomlin, et al Chemical Composition and Morphological Analysis of Atmospheric Particles from an Intensive Bonfire Burning Festival. *Environmental Science: Atmospheres*, 2, 616–633, (2022). https://doi.org/10.1039/D2EA00037G

Y. Zhou, et al. Molecular characterization of water-soluble brown carbon chromophores in seasonal snow from northern Xinjiang, China. *Environmental Science and Technology*, 56 (7), 4173-4186, (2022). doi: 10.1021/acs.est.1c07972

J.M. Tomlin, et al. Impact of Dry Intrusion Events on Composition and Mixing State of Particles During Winter ACE-ENA Study. *Atmospheric Chemistry and Physics*, 21, 18123–18146, (2021). doi: 10.5194/acp-21-18123-2021.

Y. Zhou, et al. Molecular Composition, Optical Properties, and Radiative Effects of Water-Soluble Organic Carbon in Snowpack Samples from Northern Xinjiang, China. *Atmospheric Chemistry and Physics*, 21, 8531–8555, (2021). doi: 10.5194/acp-21-8531-2021

J.M. Tomlin, et al. Chemical Imaging of Atmospheric Particles Collected from a Research Aircraft over Agricultural Fields. *ACS Earth and Space Chemistry,* 4, 2171−2184, (2020). doi: 10.1021/acsearthspacechem.0c00172

S. China, et al. Microanalysis of Primary Biological Particles Emitted from Model Grass Over its Lifecycle. *ACS Earth and Space Chemistry*, 4, 1895–1905, (2020). doi: 10.1021/acsearthspacechem.0c00144

M. Fraund, et al. Optical properties and composition of viscous organic particles found in the Southern Great Plains. *Atmospheric Chemistry and Physics*, 20, 11593–11606, (2020). doi: 10.5194/acp-20-11593-2020

S. China, et al. Rupturing Of Biological Spores As A Source Of Secondary Particles In Amazonia. *Environmental Science and Technology*, 50, 12179−12186, (2016). doi: 10.1021/acs.est.6b02896