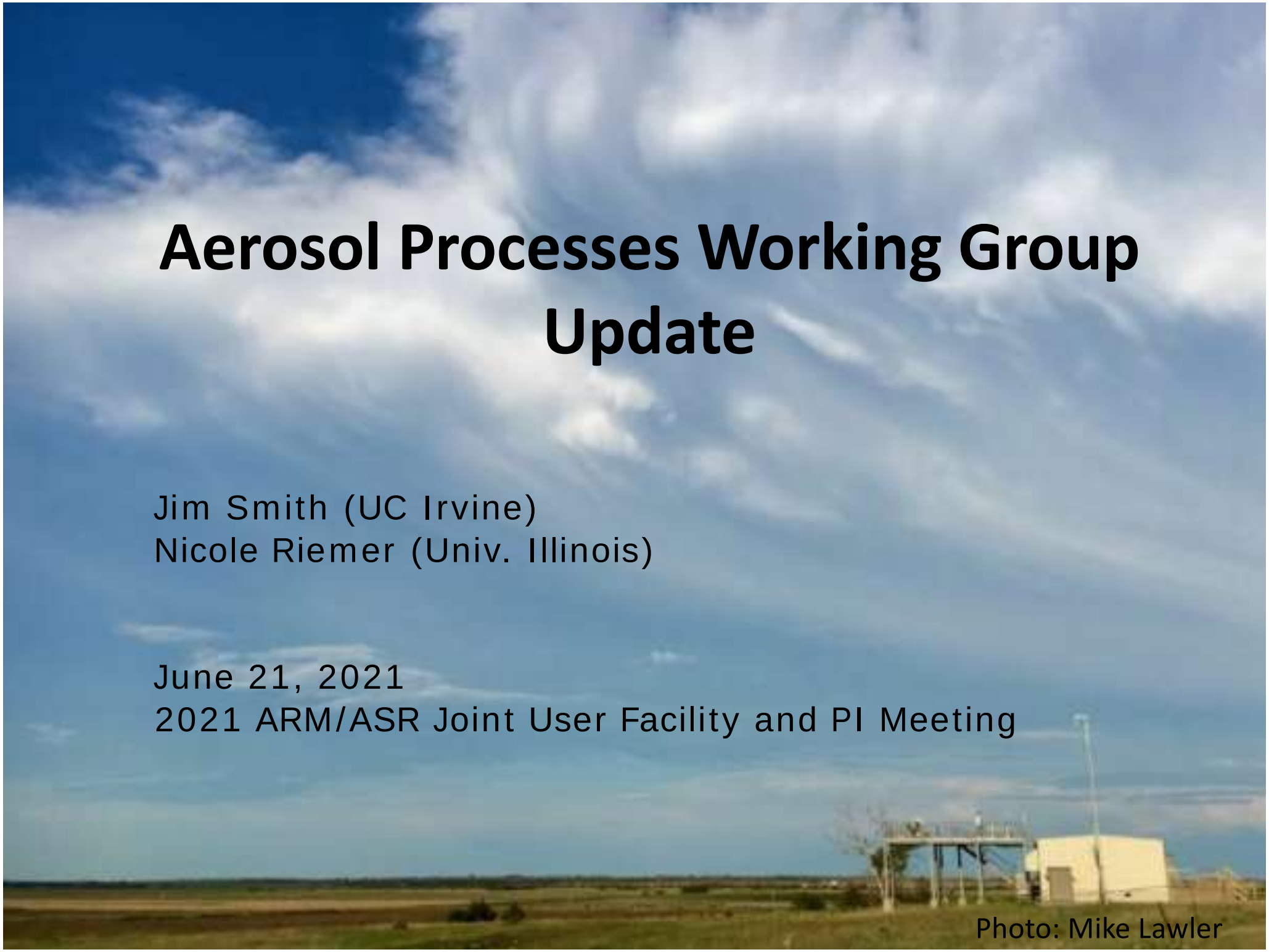
Aerosol Processes Working Group Update

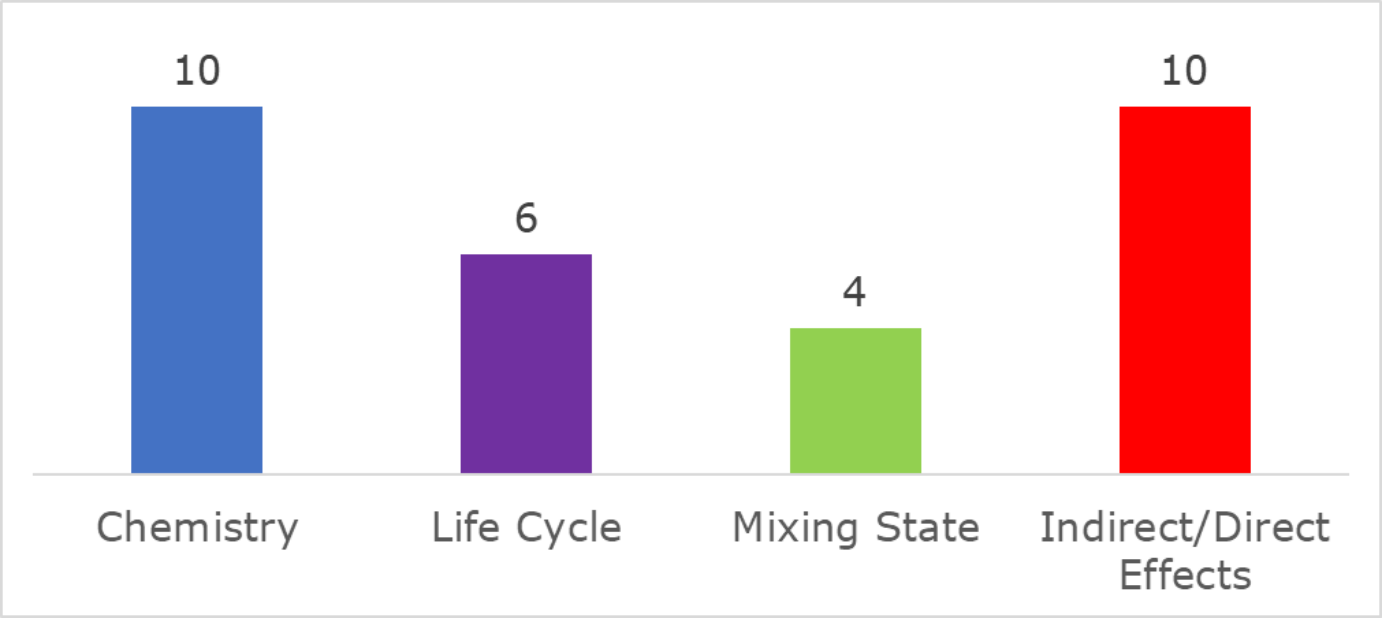
Jim Smith (UC Irvine)
Nicole Riemer (Univ. Illinois)

June 21, 2021
2021 ARM/ASR Joint User Facility and PI Meeting



Who are we?
Current research areas for Aerosol Processes WG PIs

Current # of PI-led projects by research area



Chemistry: Composition, partitioning, transformations

Life Cycle: New particle formation and particle deposition

Mixing State: Particle diversity in the ambient atmosphere and its representation in models

Direct and Indirect Effects on Climate: CCN and IN activity, scattering and absorption

What's new?
Current and future field campaigns

Eastern Pacific Cloud Aerosol Precipitation Experiment (**EPCAPE**)
2023 – 2024, San Diego, CA
Session: Tues, 11am ET

Southeast US Deployment of the AMF3 (**SEUS**)
2022 – 2027, Southeastern US

Surface Atmosphere Integrated field Laboratory (**SAIL**)
Sep 2021 – Jun 2023, Crested Butte, CO
Session: Wed, 11am ET

Tracking Aerosol Convection Interactions Experiment (**TRACER**)
Oct 2021 – Sept 2022 (IOP in summer 2022), Houston, TX

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- Can we develop better, more efficient models for SOA physico-chemical properties?
- What do laboratory and field measurements tell us about aerosol formation and evolution?

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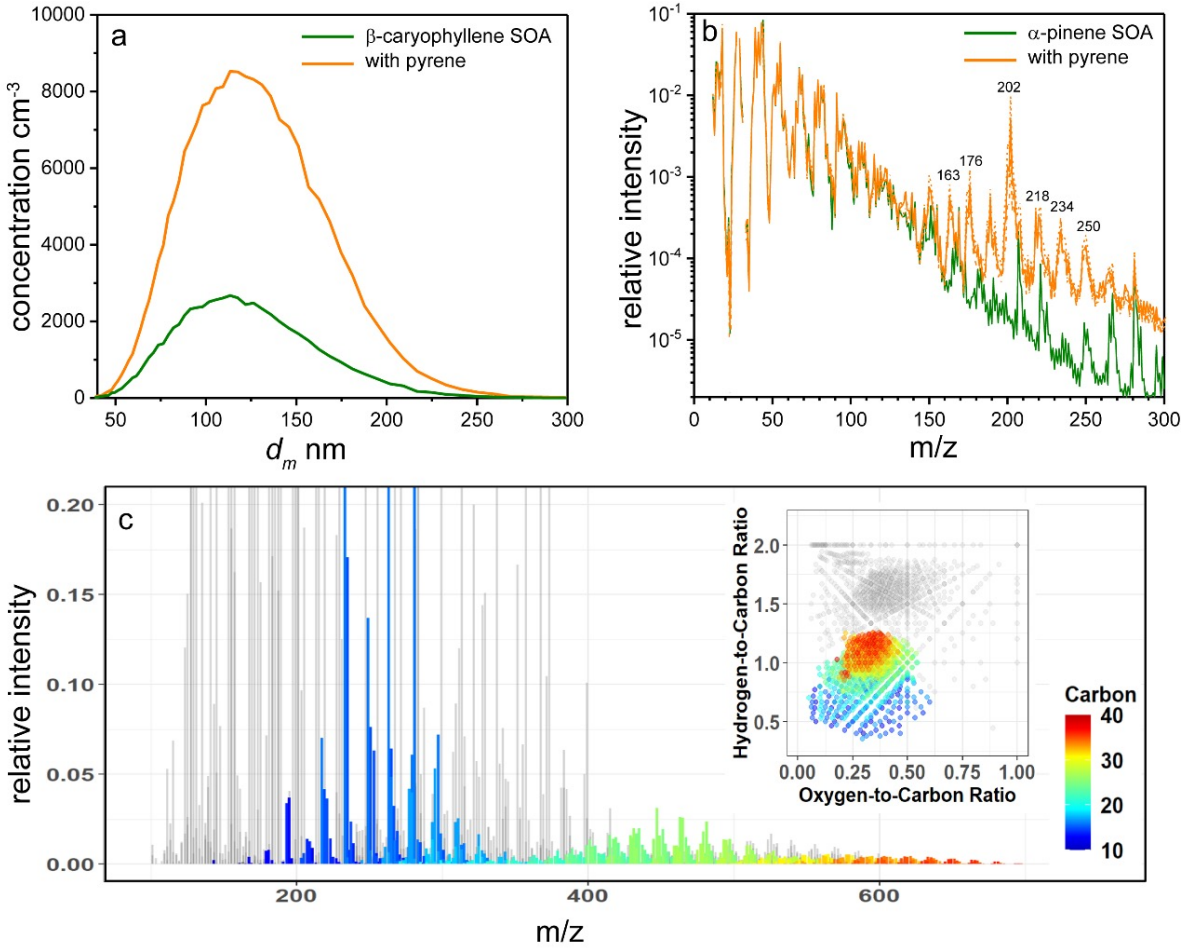
Synergy between Polycyclic Aromatic Hydrocarbons and Biogenic Secondary Organic Aerosol Particles

Objective

- Understand biogenic-anthropogenic interactions that result in higher SOA loadings and affect SOA properties

Key Findings

- The presence of PAHs during SOA formation and growth yields significantly higher particle number concentrations and mass loadings
- SOA particles formed in the presence of PAHs are more viscous and less volatile, contain more oligomers
- The "extra mass" is dominated by oxidation products of biogenic VOCs
- PAHs enhance oligomer formation



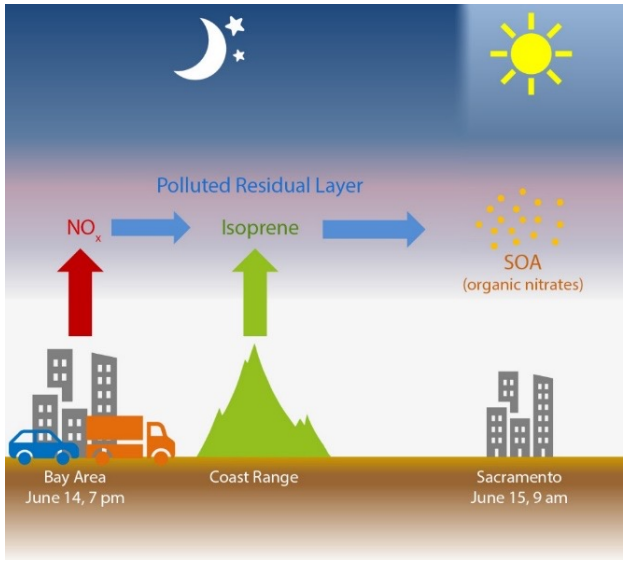
The effect of pyrene on concentration and composition of biogenic SOA. Pyrene enhances formation of oligomers containing a single oxidized pyrene and one or more oxidized α-pinene molecules (C_{16}-C_{26}-C_{36}).

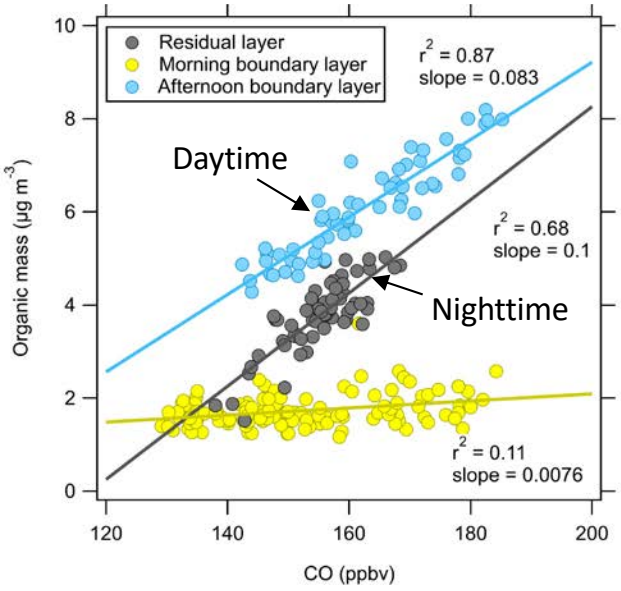
Bell, D., Suski, K., Imre, D., Shrivastava, M., Zelenyuk, A. Formation and Properties of Secondary Organic Aerosol Particles Generated by Ozonolysis of β-Caryophyllene with and without the Presence of Gas-phase Pyrene. *ACS Earth Space Chem.* 2021, *submitted;* Zelenyuk, A., Schum, S., Mazzoleni, L., Suski, K, Bell, D., Imre, D., Shrivastava, M., Kramer, A., Simonich, S. Toward Understanding the Synergetic Interactions between Polycyclic Aromatic Hydrocarbons and Biogenic Secondary Organic Aerosol Particles. *ACS Earth Space Chem.* 2021, *in preparation.*

Efficient nighttime SOA formation from anthropogenic-biogenic interactions

Objective

- Understand SOA formation from **nighttime** mixing of Bay Area pollution (NO_x) with biogenic VOCs (mainly isoprene) during the CARES field campaign





Key Finding

- Nighttime biogenic SOA (in the form of organic nitrates) was produced as efficiently as daytime SOA production in terms of $\Delta OA/\Delta CO$ ratio.

Nitrogen oxides (NOx) emissions from the Bay Area mixed with biogenic isoprene in the nighttime residual layer formed SOA at an efficiency comparable to vigorous daytime SOA formation in the same region.

Zaveri, R. A., J. E. Shilling, J. D. Fast, and S. R. Springston "Efficient nighttime biogenic SOA formation in a polluted residual layer." J. Geophysical Research, (2020), doi:10.1029/2019JD031583.

Fire and Urban Influences on SOA in the Central Amazon

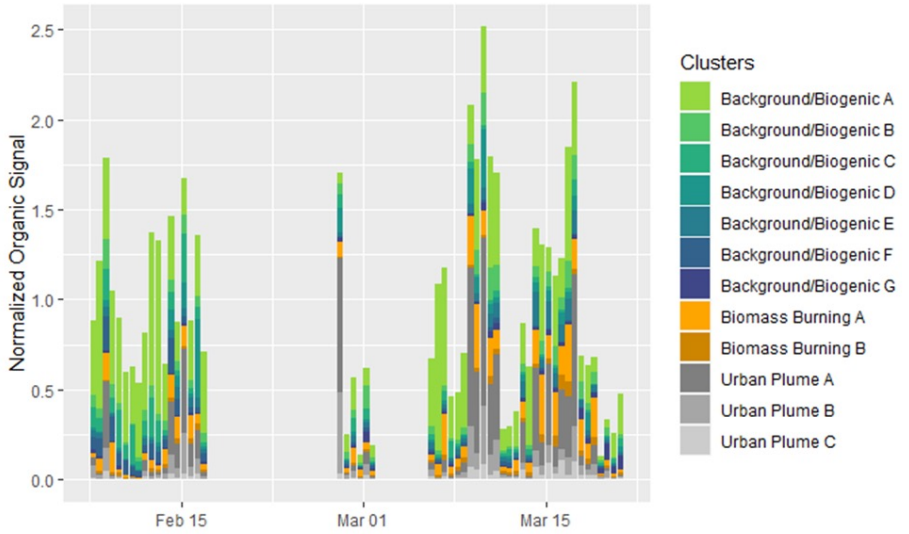
Objective

- Understand mechanisms and magnitudes of urban and biomass burning-based perturbations of secondary organic aerosol formation in the Amazon

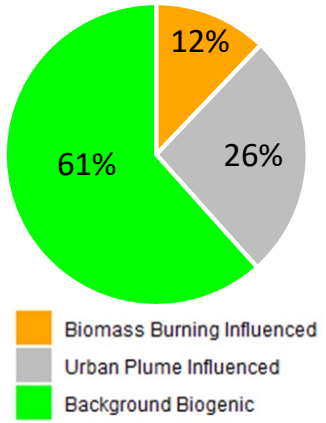
Key Finding

- Urban and fire emissions significantly contribute to SOA formation in both seasons but vary significantly both between and within seasons

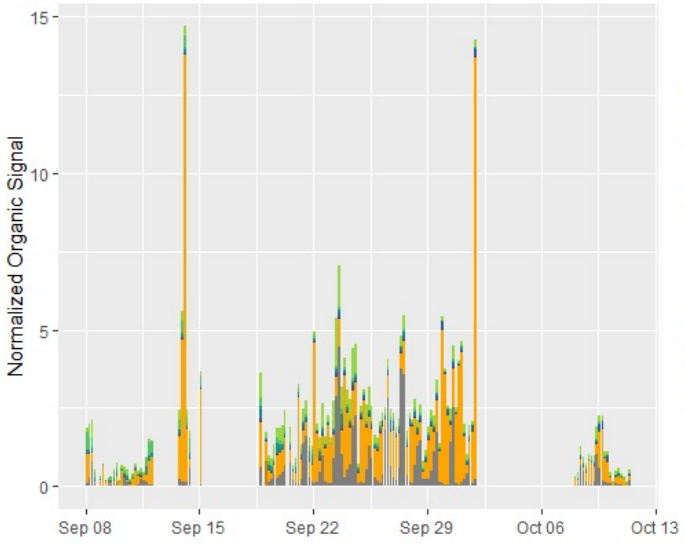
GoAmazon Wet Season



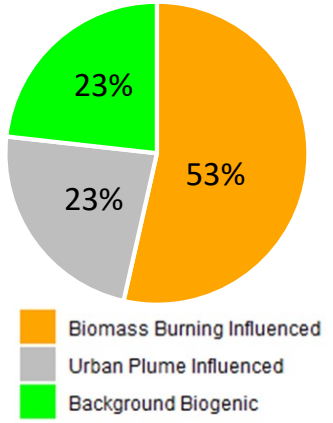
Cumulative Season Signal



GoAmazon Dry Season



Cumulative Season Signal



E. Barnes, L. Yee, R. Wernis, G. Isaacman-VanWertz, N. Kreisberg, R. Weber, S. de Sá, B. Palm, W. Hu, P. Campuzano-Jost, D. Day, A. Manzi, P. Artaxo, R. De Souza, J. Jimenez, L. Alexander, S. Martin, A. Goldstein "Chemical Signatures of Fire and Urban Influenced Secondary Aerosol Formation in the Central Amazon" *In Prep, anticipated 2021.*

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simpleSOM-MOSAIC to model SOA formation & evolution

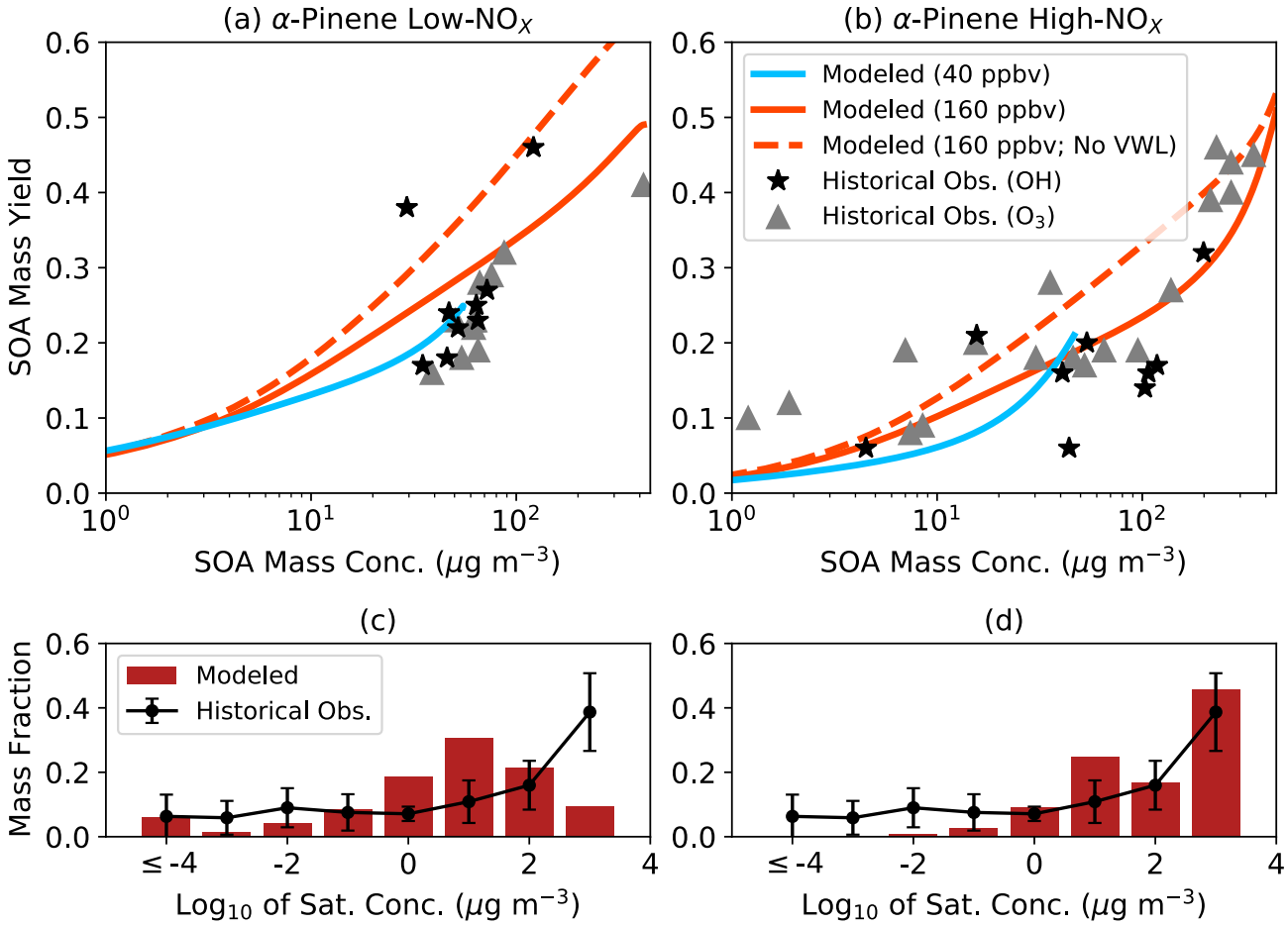
(Shantanu Jathar, Jeffrey Pierce, Christopher Cappa – DE-SC0017975)

Objective

- Develop a computationally efficient, process-level model to represent the chemistry, thermodynamic properties, and microphysics of SOA

Key Finding

- Parameters developed from one set of chamber experiments can reproduce observations of SOA mass yield, O:C, and volatility distribution gathered from other chambers



Shantanu H. Jathar, Christopher D. Cappa, Yicong He, Jeffrey R. Pierce, Wayne Chuang, Kelsey R. Bilsback, John H. Seinfeld, Rahul A. Zaveri, and Manish Shrivastava. A Computationally Efficient Model to Represent the Chemistry, Thermodynamics, and Microphysics of Secondary Organic Aerosol (simpleSOM): Model Development and Application to α-pinene SOA. Submitted to Environmental Sciences: Atmospheres.

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

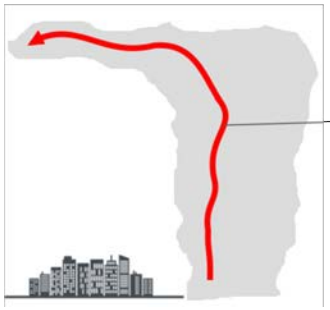
R. Bardakov, **J. A. Thornton**, I. Riipinen, R. Krejci, A. M. L. Ekman

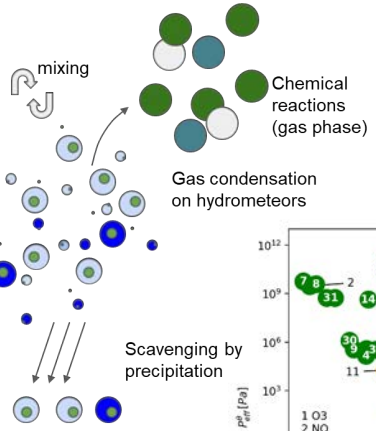
UNIVERSITY of WASHINGTON

Stockholm University

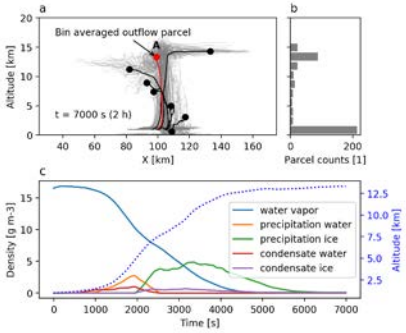
In review (Tellus)

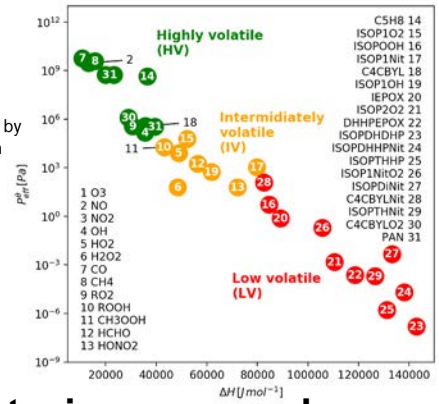
Trajectories framework





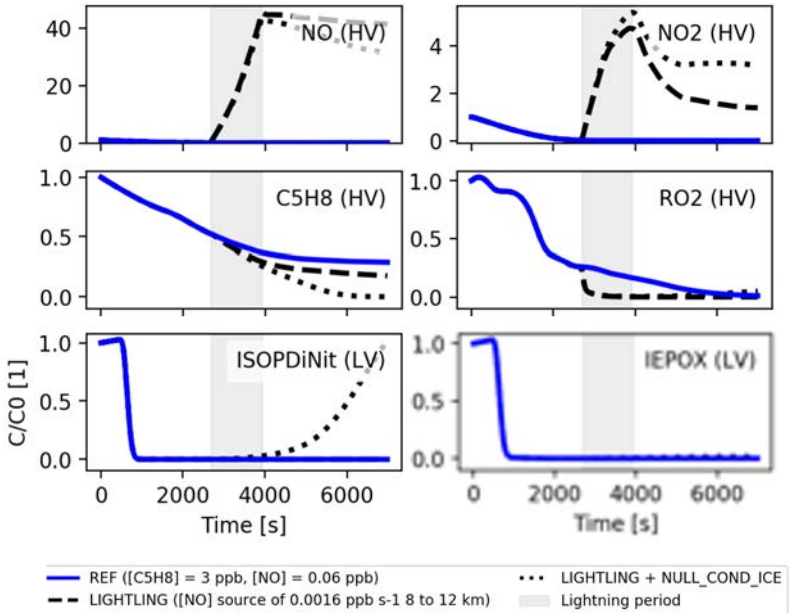
400 air parcel trajectories





31 oxidants, isoprene and oxidation products spanning wide volatility range

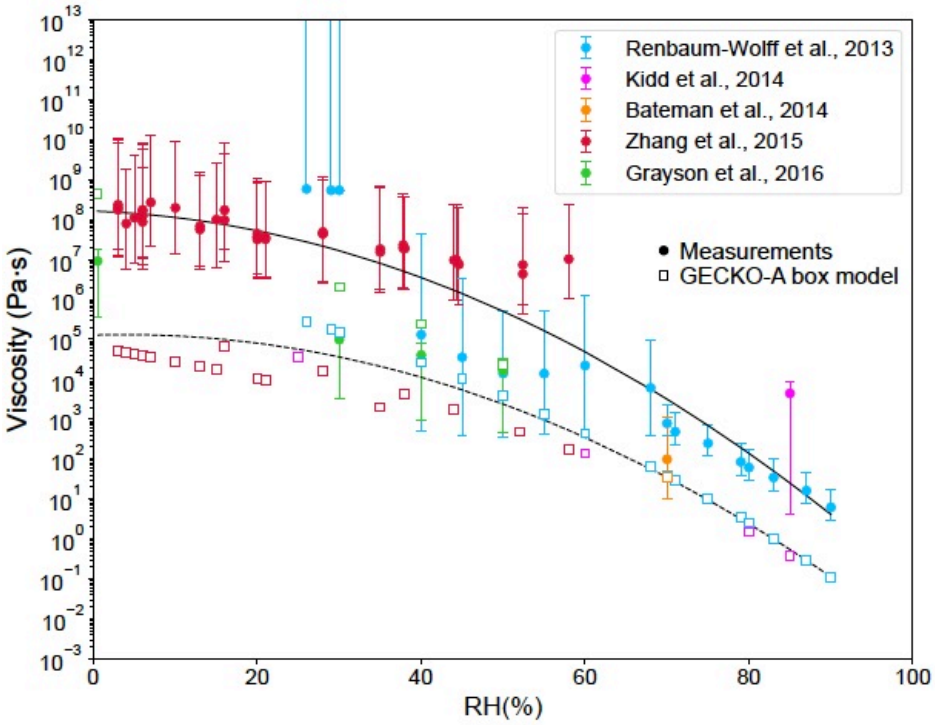
4 hydrometeor types



- 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

Viscosity of SOA & Gas-Particle Partitioning

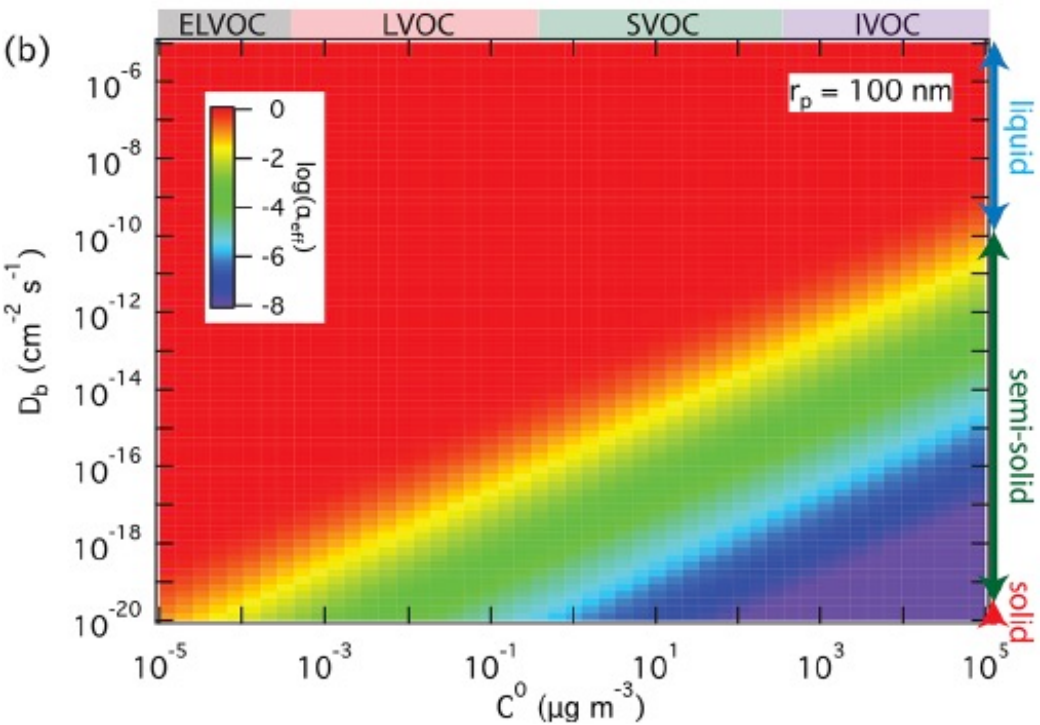
Phase State of SOA (liquid, semisolid, glassy)



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. Discuss.***, https://doi.org/10.5194/acp-2021-5117, 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.

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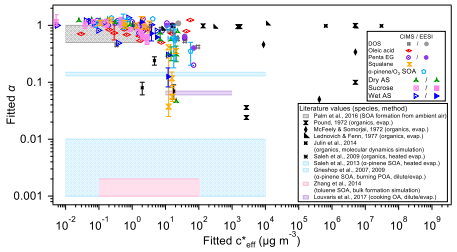
SOA Gas/Particle Partitioning: Kinetic and Equilibrium Effects

Objectives

- Quantify gas/particle mass accommodation coefficients (α) for oxidized organic compounds into a wide range of seed particle types, using a novel experimental approach for lab chambers.

Key Findings

- α ranged from 0.2 to 1 for all seeds, verified with chemically speciated measurements in both the gas and particle phases.
- Mass transfer may not be significant limitation to SOA evolution in the warmer lower troposphere.



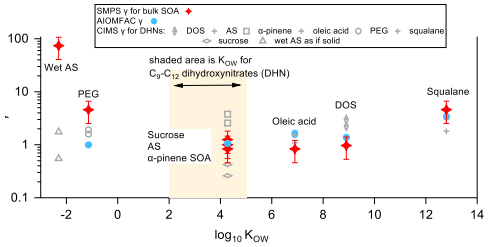
Accommodation coefficients (α) vs probe SOA molecule volatility (c) from this study and previous literature.*

Objectives

- Quantify activity coefficients (γ), a measure of non-ideal molecular interactions, for same organic gas – seed particle systems, using isothermal bulk particle evaporation and chemically speciated gases.

Key Findings

- γ increased as polarity of generated SOA molecules and pre-existing seed diverged — making SOA formation less favorable.
- Non-ideal interactions can occur in multi-component mixtures and influence SOA formation and evolution.



Activity coefficients (γ) vs seed n-octanol/water partition coeff., (K_{OW}), a metric for polarity (lipophilicility/hydrophilicity)

- X. Liu, D.A. Day, J.E. Krechmer, W. Brown, Z. Peng, P.J. Ziemann, J.L. Jimenez. Direct measurements of semi-volatile organic compound dynamics show near-unity mass accommodation coefficients for diverse aerosols. *Comms. Chem.*, 2, 98, 2019. https://doi.org/10.1038/s42004-019-0200-x
- X. Liu, D.A. Day, J.E. Krechmer, P.J. Ziemann, J.L. Jimenez. Determining Activity Coefficients of SOA from Isothermal Evaporation in a Laboratory Chamber. *Environ. Sci. Technol. Lett.*, 8, 3, 212–217, 2021. https://doi.org/10.1021/acs.estlett.0c00888.

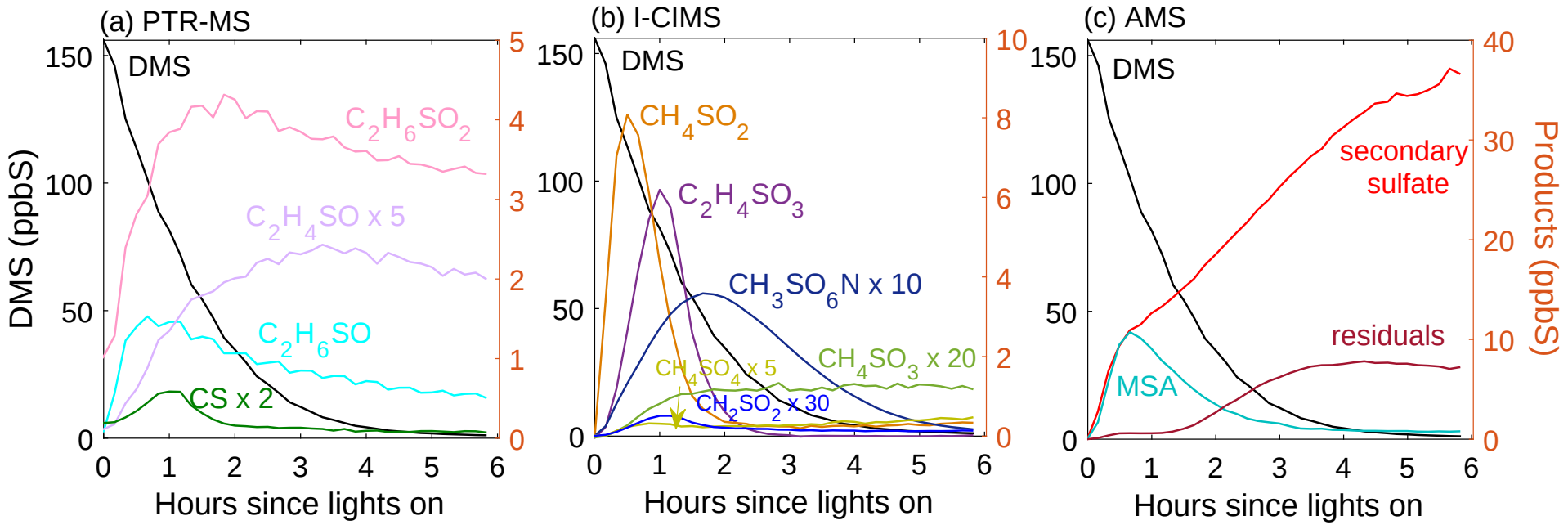
Chamber studies of dimethylsulfide (DMS) oxidation: product characterization and RO_2 chemistry

Objective

Measure the full range of S-containing products of DMS+OH, to better constrain the mechanism and rate of of sulfate aerosol formation

Key finding

Wide range of species measured in the gas and particle phases; rate of CH_3SCH_2OO isomerization measured



Qing Ye, Jesse Kroll, et al., **see poster on Thursday afternoon**

MIT ATMOSPHERIC CHEMISTRY

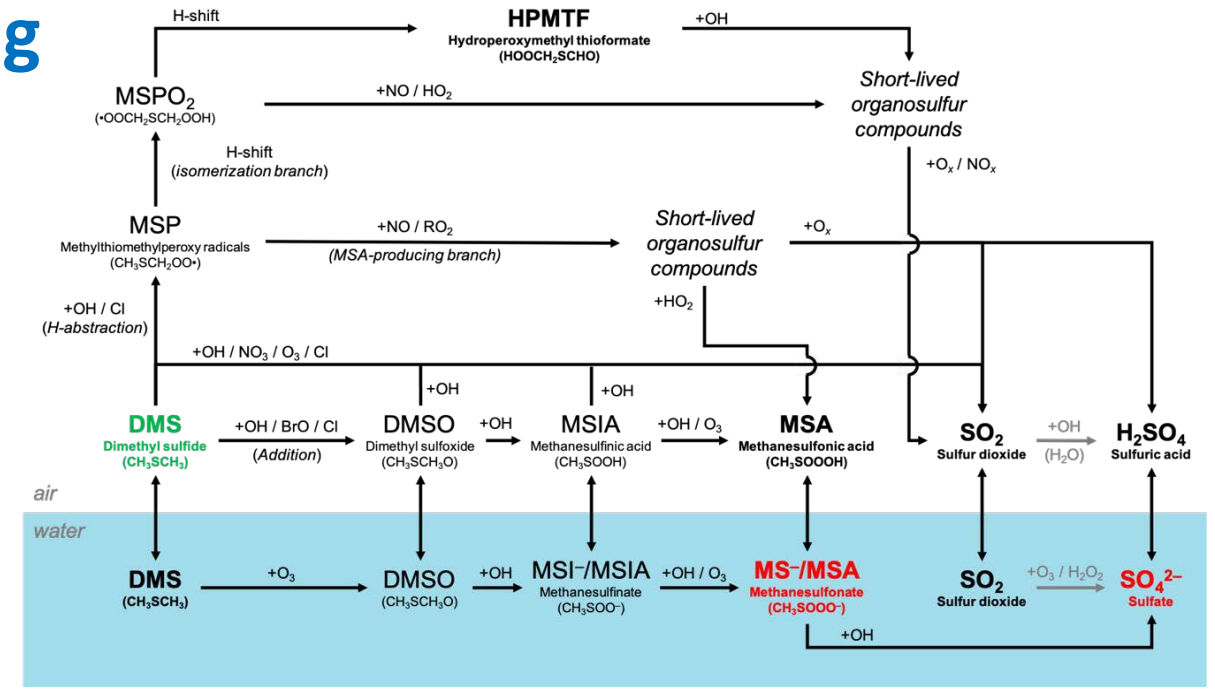
Exploring DMS oxidation and implications for global aerosol radiative forcing

Objective

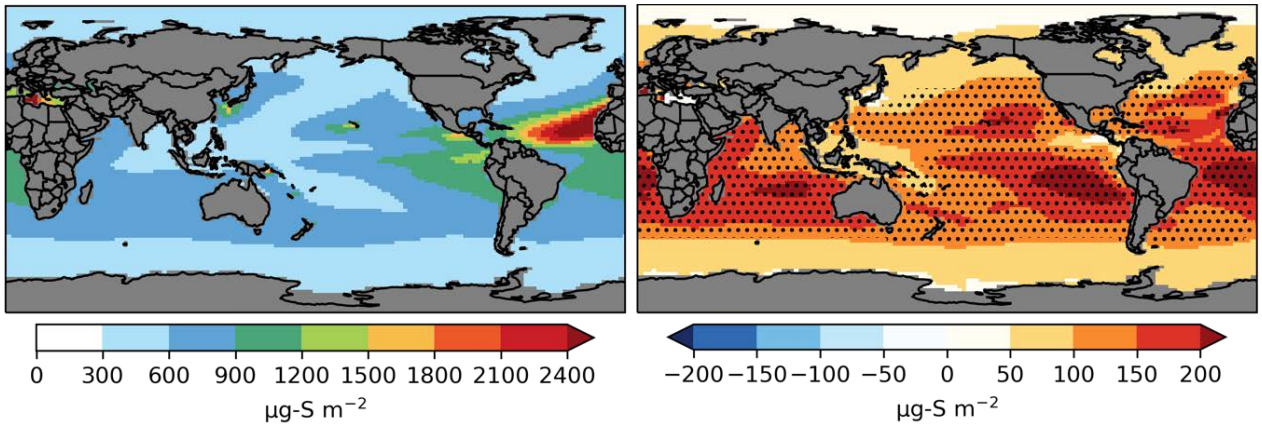
- Explore the impact of detailed DMS oxidation (including isomerization) on aerosol indirect effect

Key Finding

- Increase in pre-industrial aerosol burden dampens the aerosol indirect effect by ~25% in CESM



Pre-industrial annual mean sulfate burden (left) and increase (right) with expanded chemistry

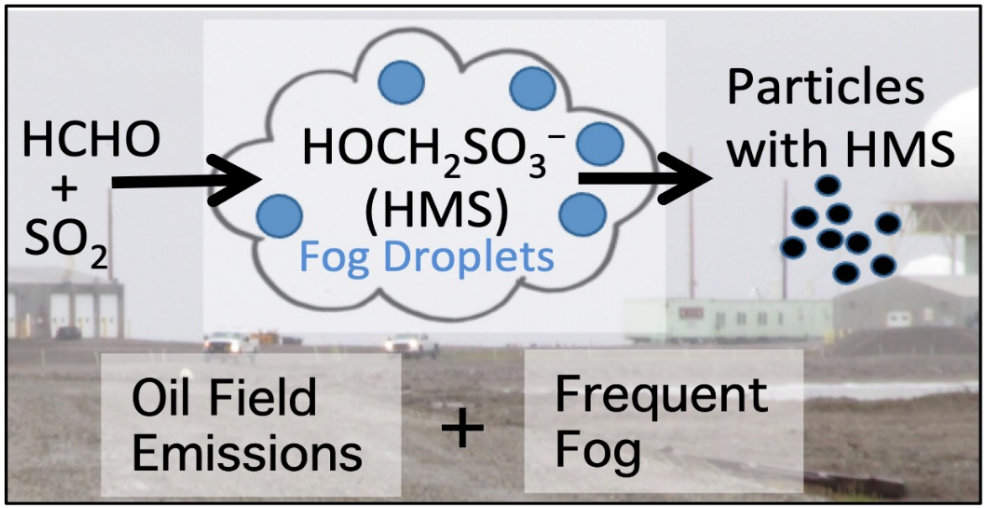


Ka Ming Fung and Colette Heald, **see poster on Thursday afternoon**

MIT ATMOSPHERIC CHEMISTRY

Aqueous Hydroxymethanesulfonate Formation in the Arctic

Kerri Pratt, Univ. of Michigan

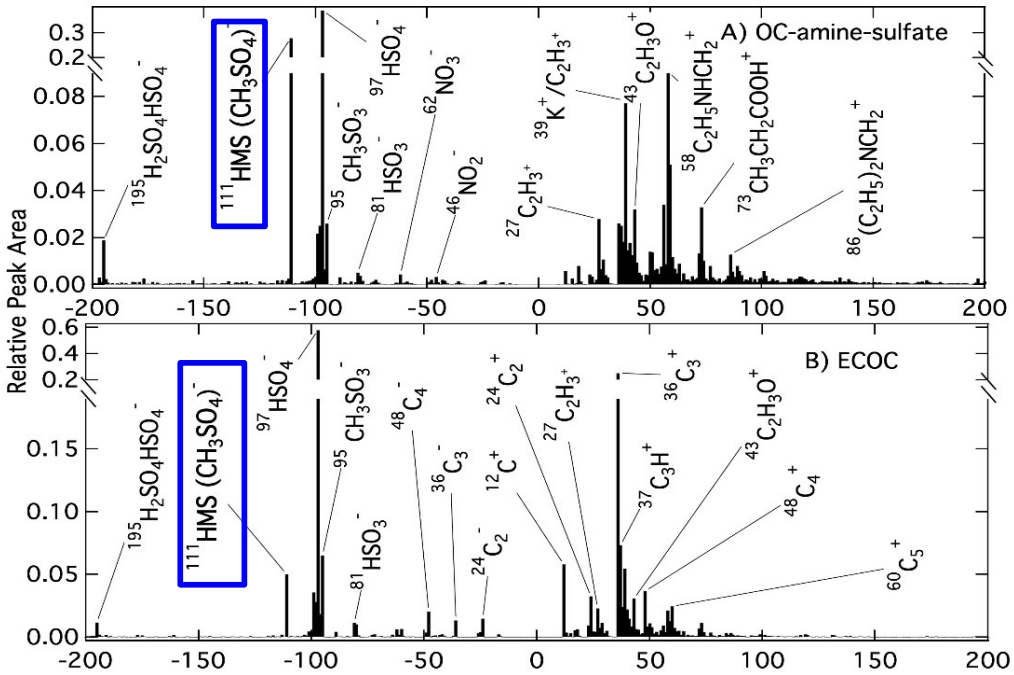


Objective

- Investigate Arctic secondary aerosol formation processes (ARM Field Campaign: Aug.-Sep. 2016 at NSA AMF3, Oliktok Point, Alaska)

Key Finding

- Hydroxymethanesulfonate (HMS) is produced during regional fog in the North Slope of Alaska oil fields from the aqueous-phase reaction of HCHO and SO_2



Single-particle mass spectra of HMS-containing particles

J. Liu, M.J. Gunsch, C.E. Moffett, L. Xu, R. El Asmar, Q. Zhang, T.B. Watson, H.M. Allen, J.D. Crounse, J. St. Clair, M. Kim, P.O. Wennberg, R.J. Weber, R.J. Sheesley, **K.A. Pratt**, "Hydroxymethanesulfonate (HMS) Formation during Summertime Fog in an Arctic Oil Field." *Under review.*

Contact: prattka@umich.edu @ArcticKerri

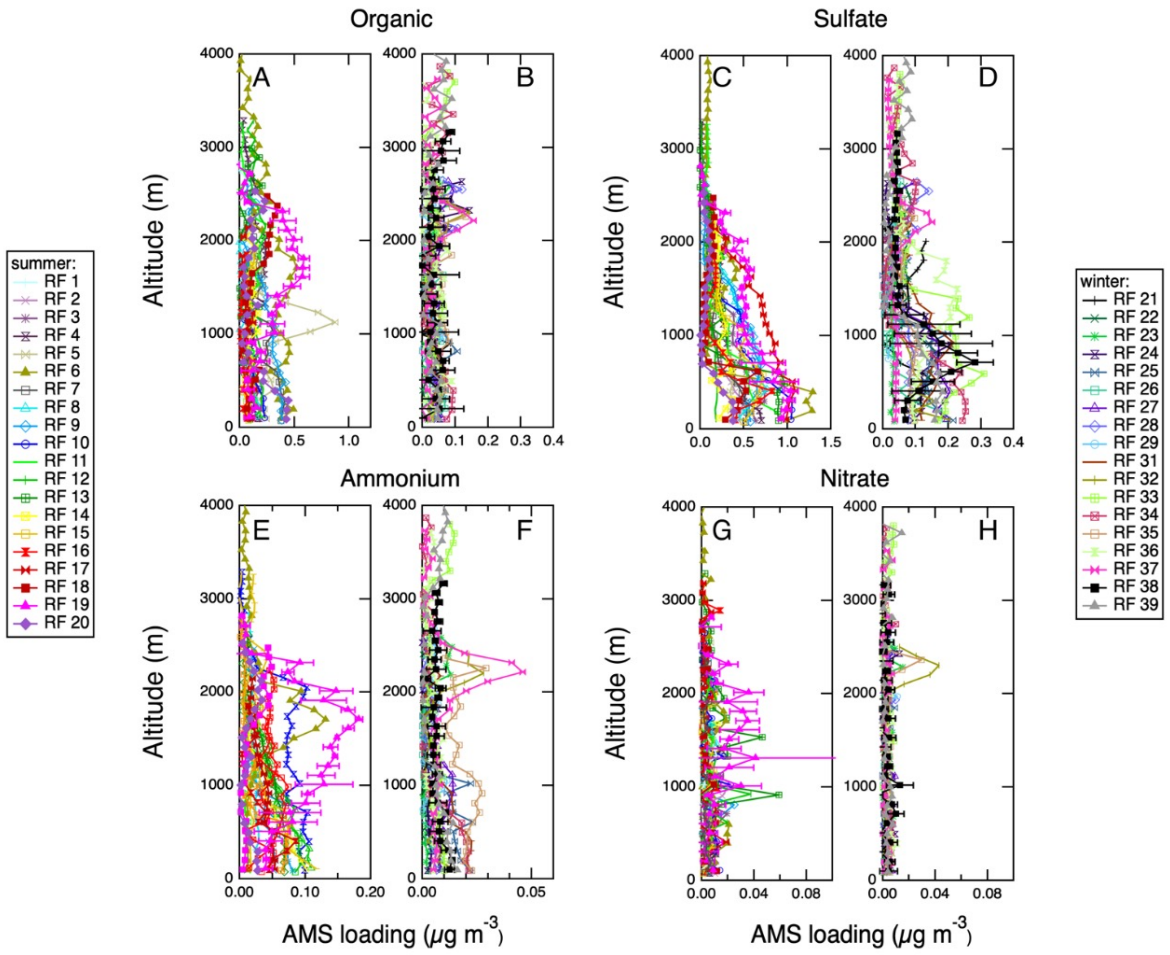
Aircraft measurements of aerosol and trace gas chemistry in the Eastern North Atlantic

Objective

- We analyze aerosol and trace gas measurements obtained by HR-TOF-AMS and PTR-MS deployed on the G-1 aircraft during the ARM ACE-ENA campaign.
- 39 research flights in the vicinity of the Graciosa Island in the Azores were carried out during two seasons, summer, 2017 and winter, 2018.

Key Finding

- The Eastern North Atlantic region was found to be very clean, with average non-refractory sub-micrometer aerosol mass loading of 0.6 μg m^{-3} in the summer and 0.1 μg m^{-3} in the winter. The aerosol chemical composition was dominated by sulfate and highly processed organics.



Vertical profiles of organic, sulfate, ammonium and nitrate, segregated by flight. Examination of vertical profiles of aerosol and gas chemistry during ACE-ENA reveals an interplay of local marine emissions and long-range transported aged aerosol.

Maria A. Zawadowicz, Kaitlyn Suski, Jiumeng Liu, Mikhail Pekour, Jerome Fast, Fan Mei, Arthur J. Sedlacek, Stephen Springston, Yang Wang, Rahul A. Zaveri, Robert Wood, Jian Wang, John E. Shilling. Aircraft measurements of aerosol and trace gas chemistry in the Eastern North Atlantic. *Atmospheric Chemistry and Physics* (accepted), https://doi.org/10.5194/acp-2020-887, 2021.

Aerosol Life Cycle: What are the questions?

- What processes control the formation of new particles in the boundary layer?
- Does new particle formation occur aloft and, if so, where do newly formed particles go?
- What processes control aerosol particle deposition?

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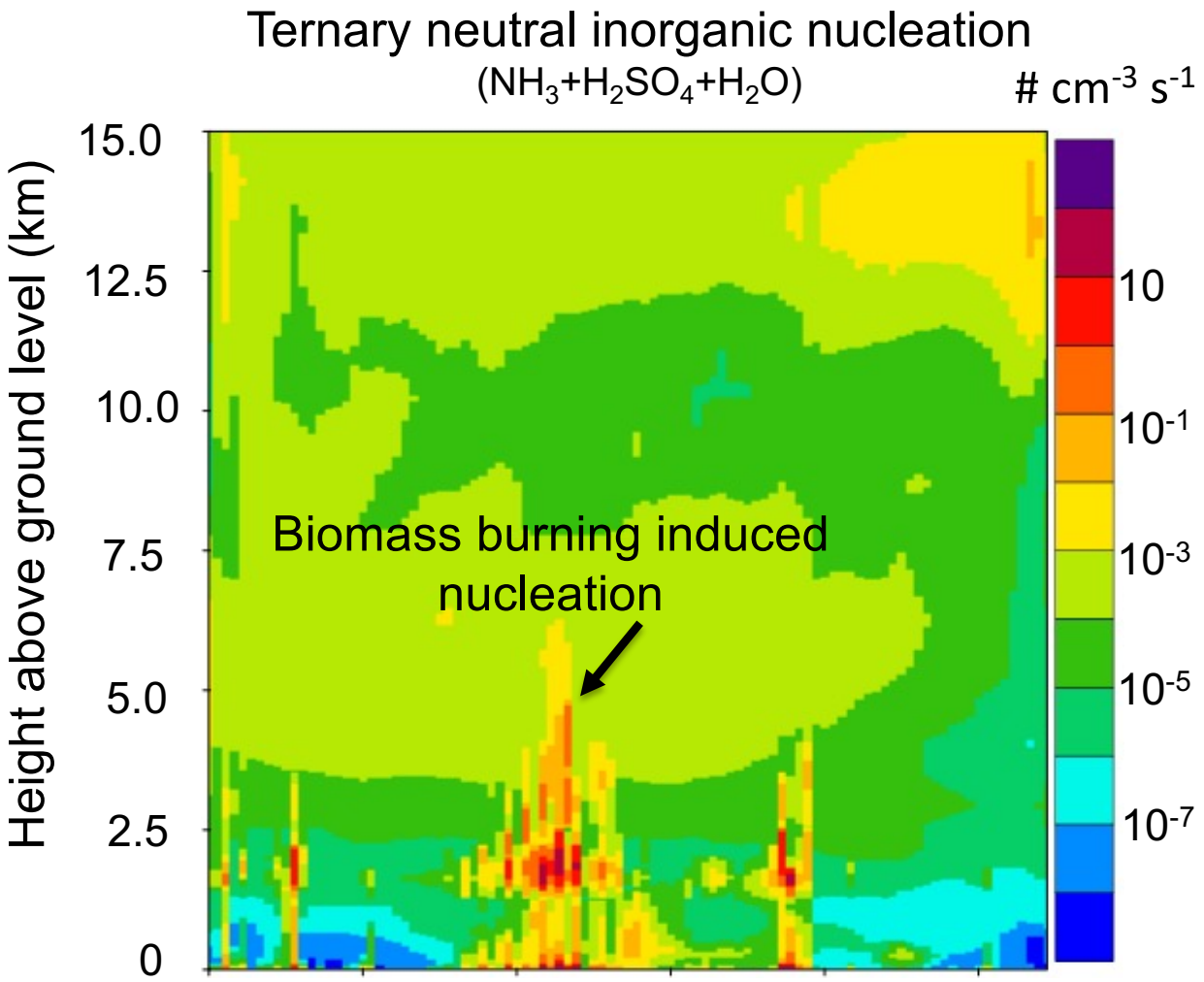
New particle formation in biomass burning plumes

Objective

- Understand new particle formation and ultrafine particle composition in biomass burning plumes

Key Finding

- Biomass burning induces nucleation and formation of large number of ultrafine particles, especially from ternary-neutral inorganic mechanism over the Amazon



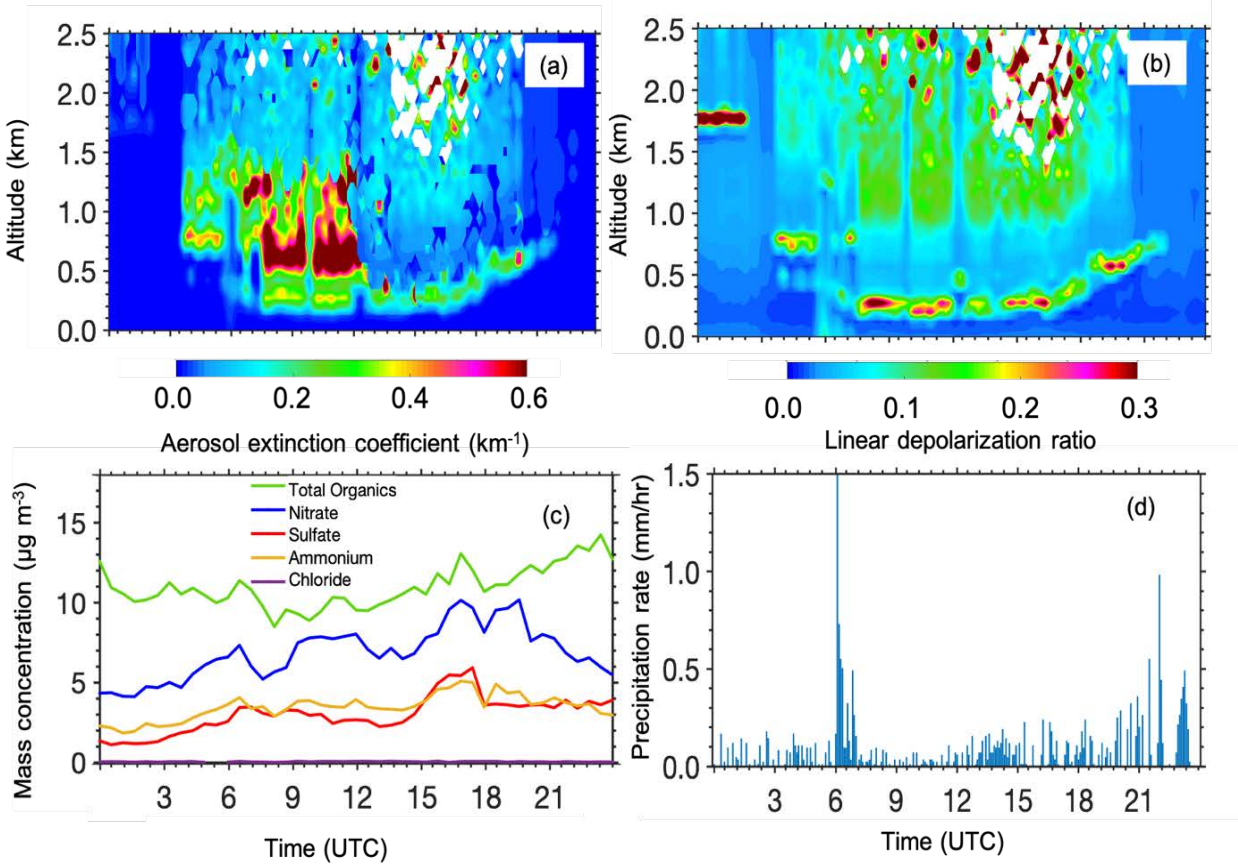
Simulated zonal mean vertical distribution of nucleation rates over the Amazon showing large new particle formation in biomass burning plumes upto 5 km altitude with plume rise

M. Shrivastava, Q. Rasool, B. Zhao, B. Gaudet, Y. Liu, M.O. Andreae, C. Schulz, J. Schneider, S. Jathar, A. Akherati, J. Shilling et al. Impact of wildfires on new particle formation over the Amazon rainforest, Manuscript in preparation

Identification of Biological Aerosol Particle Events

Objective: To use existing ARM data to identify possible instances of pollen and fungal spore primary biological particle events

Key Finding: Using lidar depolarization, extinction coefficients, aerosol composition and precipitation, we identify 6-15 days/yr as pollen events and 4-11 days/yr with pollen rupture events, with 16-33 days/yr with fungal spore rupture events



Subba, T., M.J. Lawler and A.L. Steiner, Estimation of possible primary biological particle emissions and rupture events at the Southern Great Plains ARM site*, JGR-Atmospheres*, in review.

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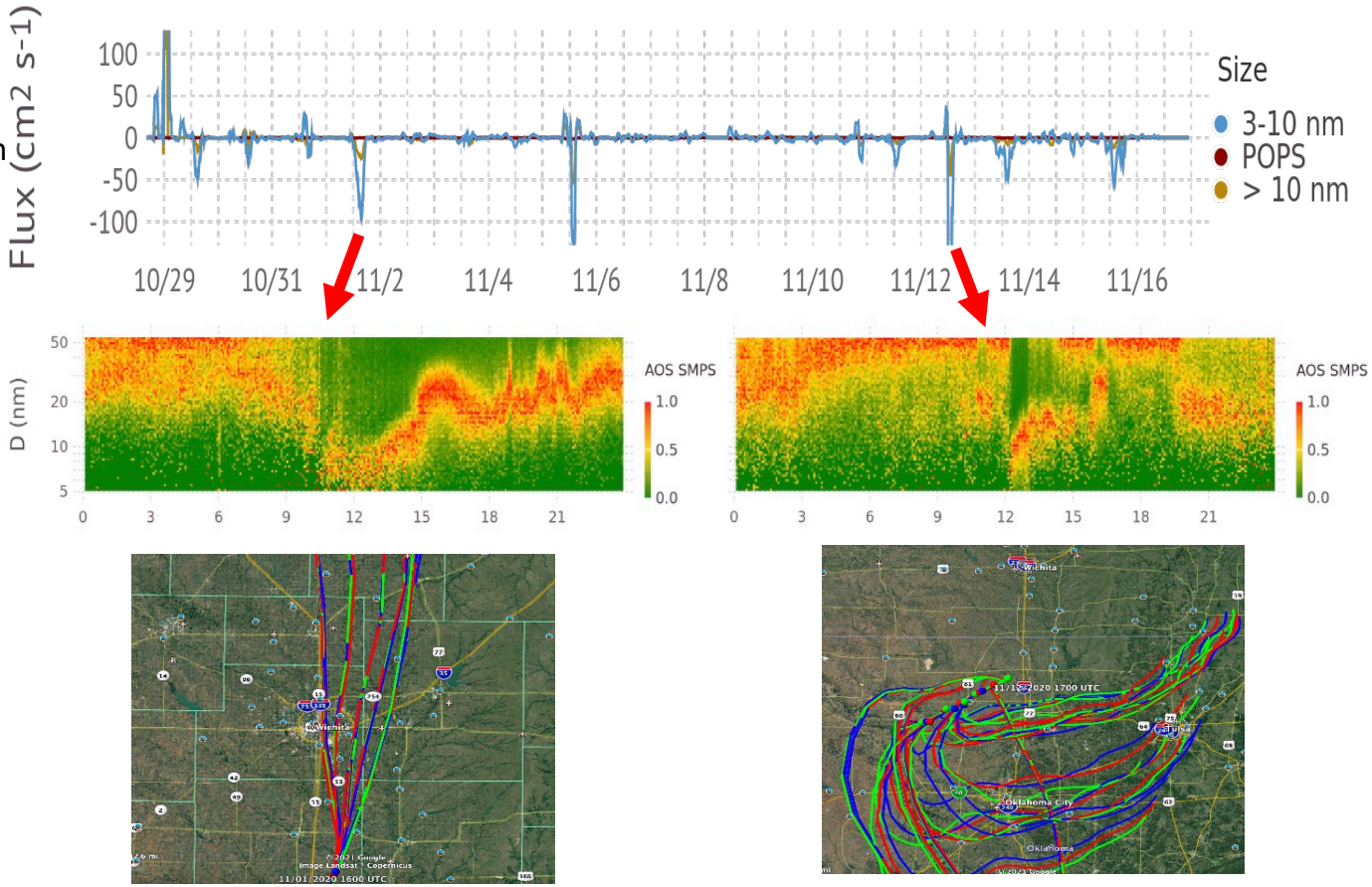
Vertical turbulent fluxes of freshly nucleated particles at the DOE ARM SGP site

Objective

- Measure vertical turbulent fluxes of >3 nm and >10 nm sized particles for separation of near-surface and residual-layer new particle formation (NPF) events

Key Findings

- Detected seven events with an over an order of magnitude increase in 3 to 10 nm-sized particle number concentrations
- Most small particle events coincided with the downward flux of 3-10 nm size particles, suggesting that formation occurred aloft
- Downward fluxes were observed when airmass trajectories passed over either Tulsa, Oklahoma City, or Wichita prior to arrival
- 10-50 nm particles had kappa values < ~0.2, suggesting that organic compounds dominated the sub-50 nm size particle chemical composition



The 3 to 10 nm-sized particle vertical turbulent fluxes, SMPS, and back trajectories

Md Maksimul Islam, N., Meskhidze, and M. Petters, "Vertical turbulent fluxes of freshly nucleated particles at the DOE ARM SGP site", in preparation.

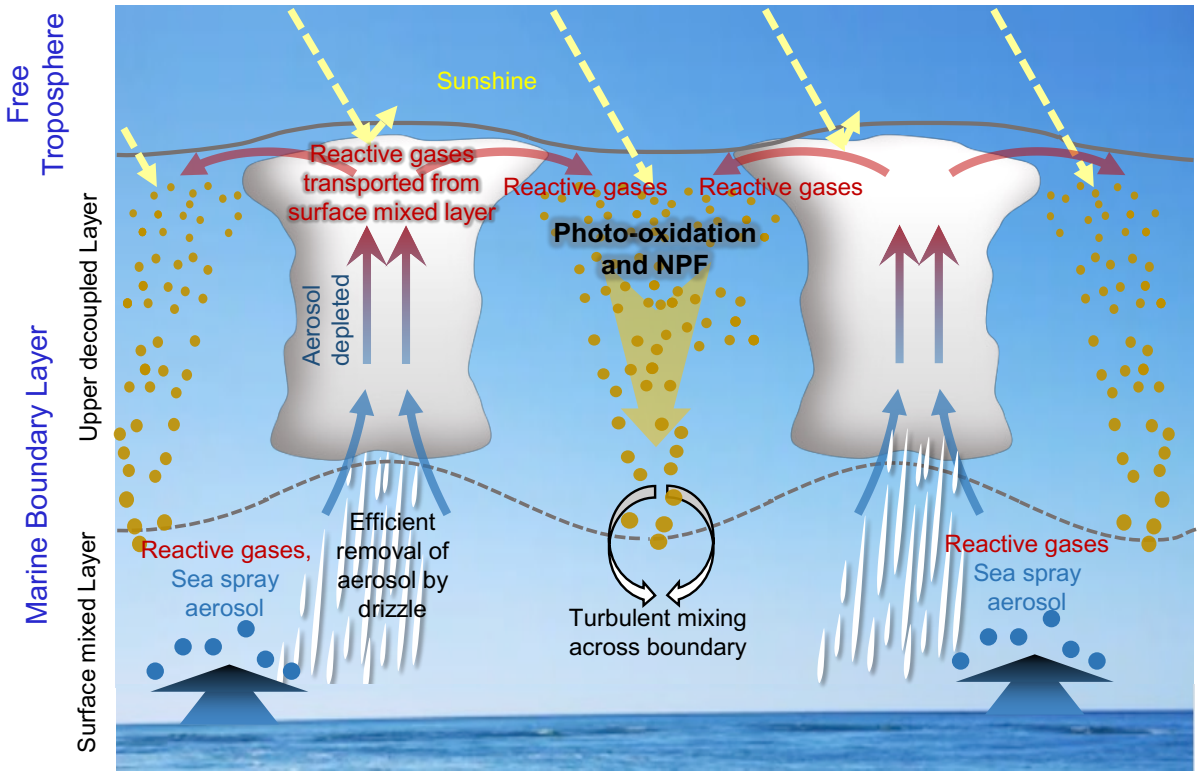
New Particle Formation in the Remote Marine Boundary Layer over Mid-latitude Oceans

Objective

- To understand the source of nucleation mode particles in remote marine boundary layer

Key Finding

- New particle formation occurs frequently in the upper part of remote marine boundary layer over mid-latitude oceans following the passage of cold fronts.
- The newly formed particles subsequently grow and can contribute substantially to cloud condensation nuclei in the remote marine boundary layer and thereby impact marine low clouds.



The new particle formation in the upper marine boundary layer is facilitated by a combination of efficient removal of existing particles by precipitation, cold air temperatures, vertical transport of reactive gases from the ocean surface, and high actinic fluxes in a broken cloud field following the passage of cold fronts.

Zheng, G. J., Y. Wang, R. Wood, M. P. Jensen, C. A. Kuang, I. L. McCoy, A. Matthews, F. Mei, J. M. Tomlinson, J. E. Shilling, M. A. Zawadowicz, E. Crosbie, R. Moore, L. Ziemba, M. O. Andreae, and J. Wang. 2021. "New particle formation in the remote marine boundary layer." *Nature Communications* **12**(1), doi: 10.1038/s41467-020-20773-1.

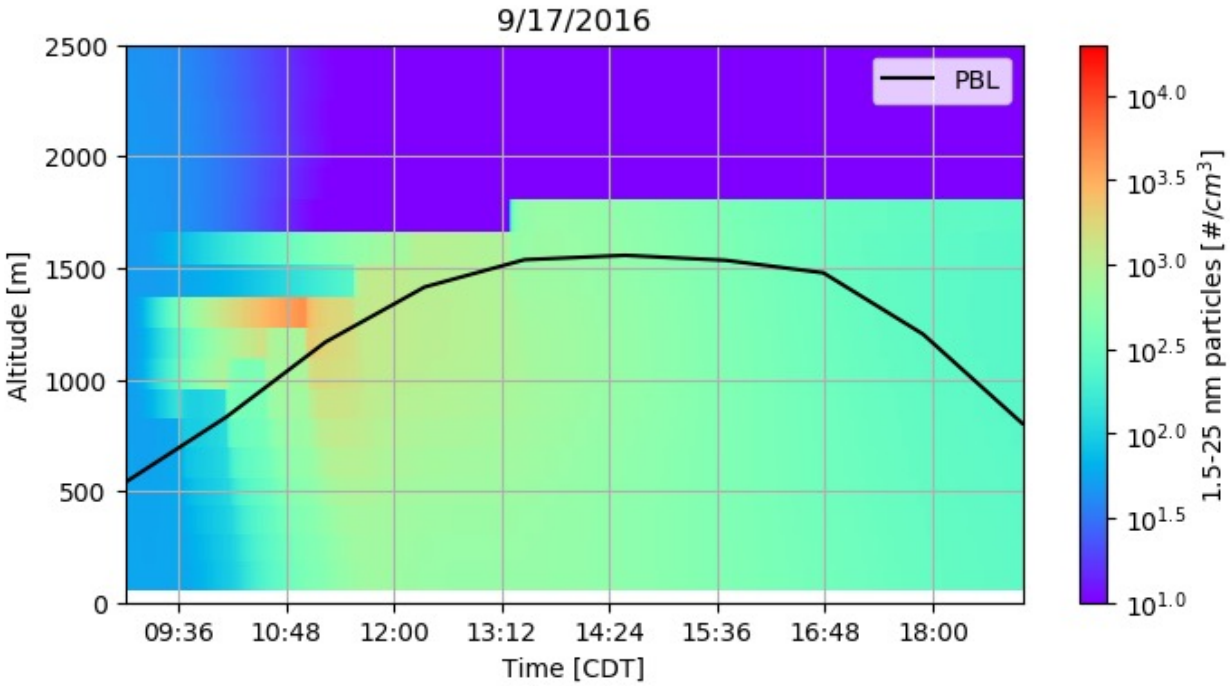
Vertically resolved new-particle formation at SGP

Objective

- Understand the vertical profile of new-particle formation/growth in and near the boundary layer at SGP.
- Use surface and airborne measurements from the HI-SCALE campaign with a 1D column version of the SOM-TOMAS aerosol chemistry-microphysics model.

Key Finding

- Inhomogeneities in precursor vapors and temperature lead to nucleation/growth initiating near the mixed-layer top with delayed mixing to the surface.



Timeseries of the vertical profile of nucleation-mode (1-25 nm) aerosol number concentrations as simulated by SOM-TOMAS. The simulation agrees with surface observations in timing, size, and concentrations. On this day, nucleation initiates in the residual boundary layer due to enhanced SO_2 (measured by aircraft) and lower temperatures; the newly formed particles entrain into the mixed layer 1-2 hours later.

Samuel E. O'Donnell, James N. Smith, Ali Akherati, Yicong He, John E. Shilling, Fan Mei, John Hubbe, Stephen R. Springston, Joel Thornton, Emma L. D'Ambro, Ben H. Lee, Siegfried Schobesberger, Michael J. Lawler, Chuck Long, Chongai Kuang, Jerome Fast, Anna Hodshire, Shantanu Jathar, and Jeffrey R. Pierce, "Process level modeling of vertically resolved new-particle formation at the Southern Great Plains observatory." *In preparation*

Aerosol Life Cycle: What are the questions?

- What processes control the formation of new particles in the boundary layer?
- Does new particle formation occur aloft and, if so, where do newly formed particles go?
- What processes control aerosol particle deposition?

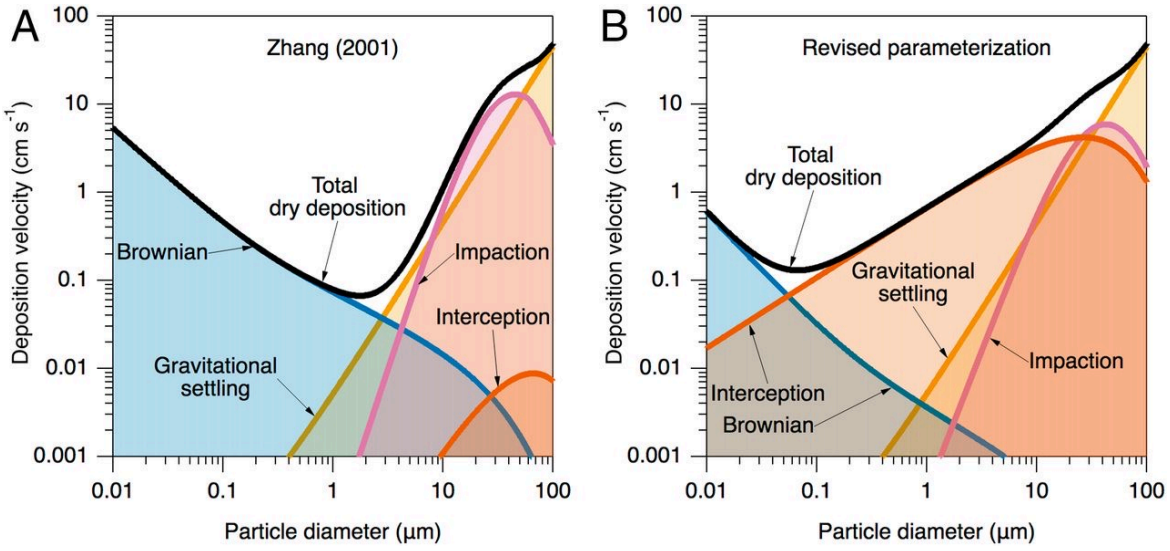
Improved understanding of size-resolved particle deposition

Objective

- Understand what processes control particle deposition, and improve their representation in models

Key Finding

- Field observations suggest that interception is underestimated in current model representations of deposition; revising particle dry deposition changes model estimates of size-resolved aerosol lifetime, mass loadings, and direct and indirect effects



Comparison of processes in revised parameterizations, highlighting the enhanced role of interception, coupled to a reduce role for gravitational settling. These changes shift the size-resolved deposition velocity.

E.W. Emerson, A.L. Hodshire, H. M. DeBolt, K.R. Bilsback, J.R. Pierce, G.R. McMeeking, D.K. Farmer. Revisiting particle dry deposition and its role in radiative effect estimates. Proceedings of the National Academy of Sciences. 2020, 117 (42) 26076-26082; DOI: 10.1073/pnas.2014761117

Mixing State: What are the questions?

- What is the aerosol mixing state of the ambient aerosol in different environments?

- How well can we represent mixing state in regional and global models?

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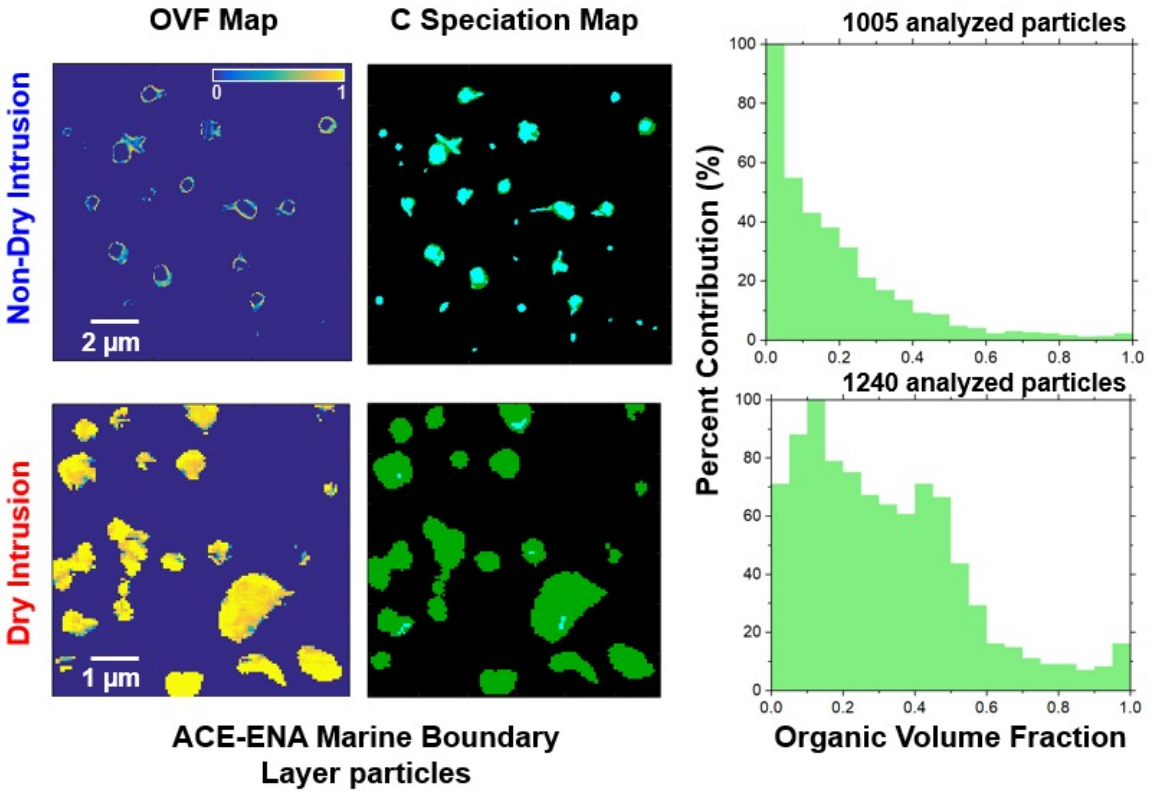
Impact of Dry Intrusion Events on Composition and Mixing State of Particles

Objective

- To investigate the physiochemical properties of individual atmospheric particles in the Eastern North Atlantic during long-range transport episodes

Key Finding

- Based on a chemical imaging analysis, carbonaceous particles were identified as the dominant particle-type in the region with significant enhancements of inorganic components (ammonium nitrate/sulfate) during dry intrusion (DI) periods.
- **Organic volume fraction (OVF) during DI events increased by a factor of 2.5 and 1.25 in the MBL and FT**, respectively, indicating that entrainment of particles from long-range continental sources alters particle-types present in MBL.



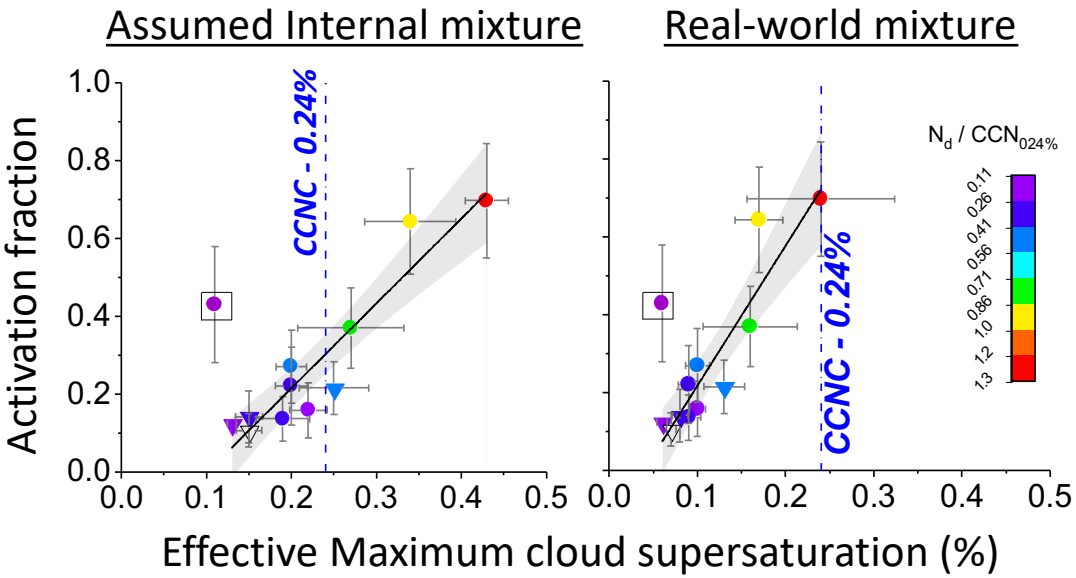
Representative Organic Volume Fraction (OVF) and Carbon speciation maps and corresponding histograms showing fractions of particles with different OVF values. Upper and lower plots contrast organic/inorganic composition and mixing states of particles from DI atmospheric transport episodes identified in ACE-ENA study.

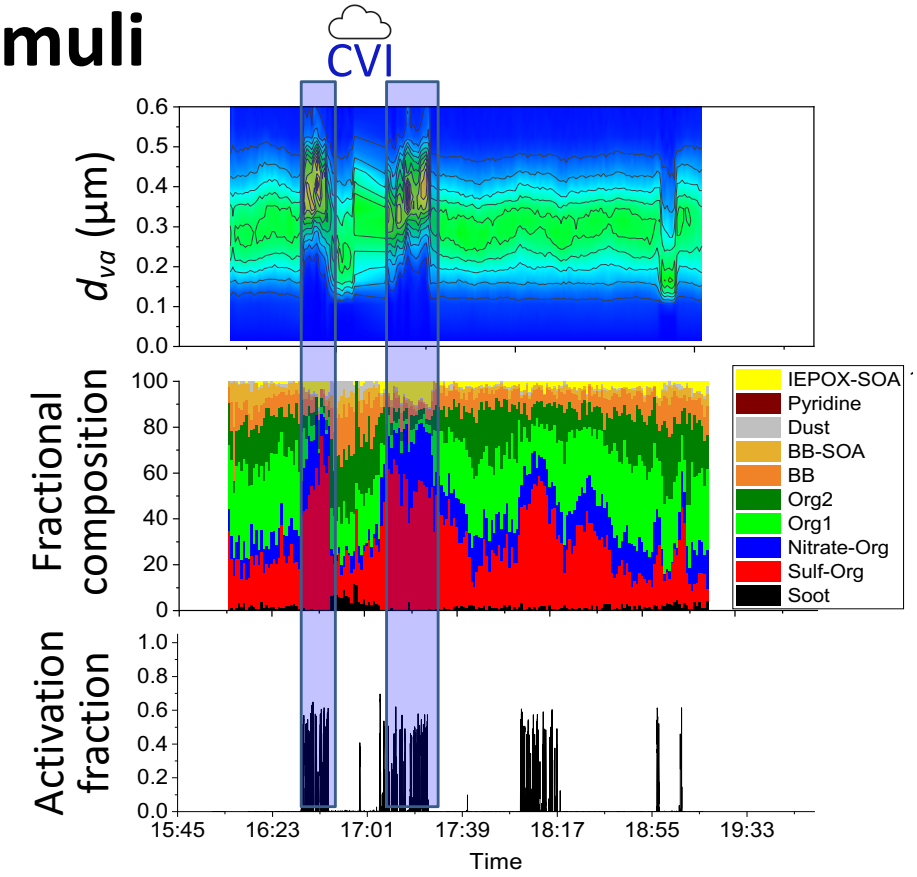
J.M. Tomlin, K.A. Jankowski, D.P. Veghte, S. China, P. Wang, M. Fraund, J. Weis, G. Zheng, Y. Wang, F. Rivera-Adorno, S. Raveh-Rubin, J.E. Shilling, D.A. Knopf, R.C. Moffet, M.K. Gilles, J. Wang, A. Laskin. Impact of Dry Intrusion Events on Composition and Mixing State of Particles During Winter ACE-ENA Study. *Atmospheric Chemistry and Physics*, 2021, submitted.

Real-World Aerosol Mixing State Necessary to Represent Activation Fraction of Shallow Cumuli

Objective

- Understand aerosol-cloud interactions in shallow cumuli *using single particle measurements*





Flight averaged effective supersaturation calculated using below-cloud aerosol composition and size distributions for assumed internal mixture vs. measured real-world mixing state

Time series for vacuum aerodynamic size distribution (top), number fraction of aerosol class (middle), and cloud activation fraction (bottom) for 04/25/2016

Key Findings

- Cloud residuals are larger and more hygroscopic than below-cloud aerosol
- Assuming internal mixtures overestimates effective cloud supersaturations by a factor of 2

G. Saliba, D. M. Bell, K. J. Suski, J. Shilling, F. Mei, G. Kulkarni, A. C. Varble, J. H. Muelmenstaedt, J. Wang, J. A. Thornton, J. Tomlinson, J. Fast, A. Zelenyuk. Aircraft measurements of single particle size and composition reveals real-world mixing state is necessary to explain activation fraction during HI-SCALE (*submitted*)

Regime-dependent Mixing State of Black Carbon Particles Produced by Biomass Burn Events

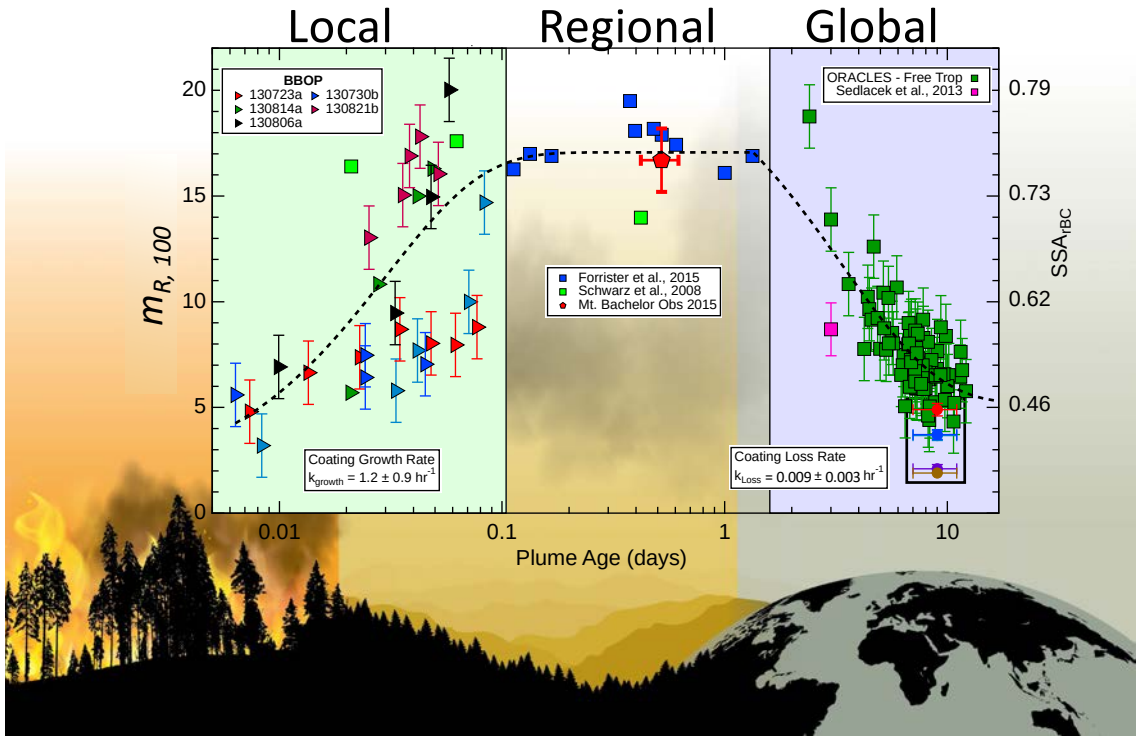
Objective

- Examine of the BC mixing state (i.e., ratio of coating to core for 100 nm BC cores; $m_{R,\,100}$) to probe the lifecycle of biomass burn black carbon (BB-BC) particles.

Key Finding

- Using datasets collected during BBOP, LASIC and ORACLES (NASA), our analysis suggests that the lifecycle of biomass burning aerosols can be partitioned into ***three spatial/temporal regimes: local, regional, and global*** as captured by three distinct phases of mixing state during particle aging.

Change in $m_{R,100}$ yields rate information on SOA production



- Coating growth continues in the local regime until SOA is exhausted.
- Little change in $m_{R,100}$ in regional regime.
- Coating material is lost through oxidative losses in global regime.

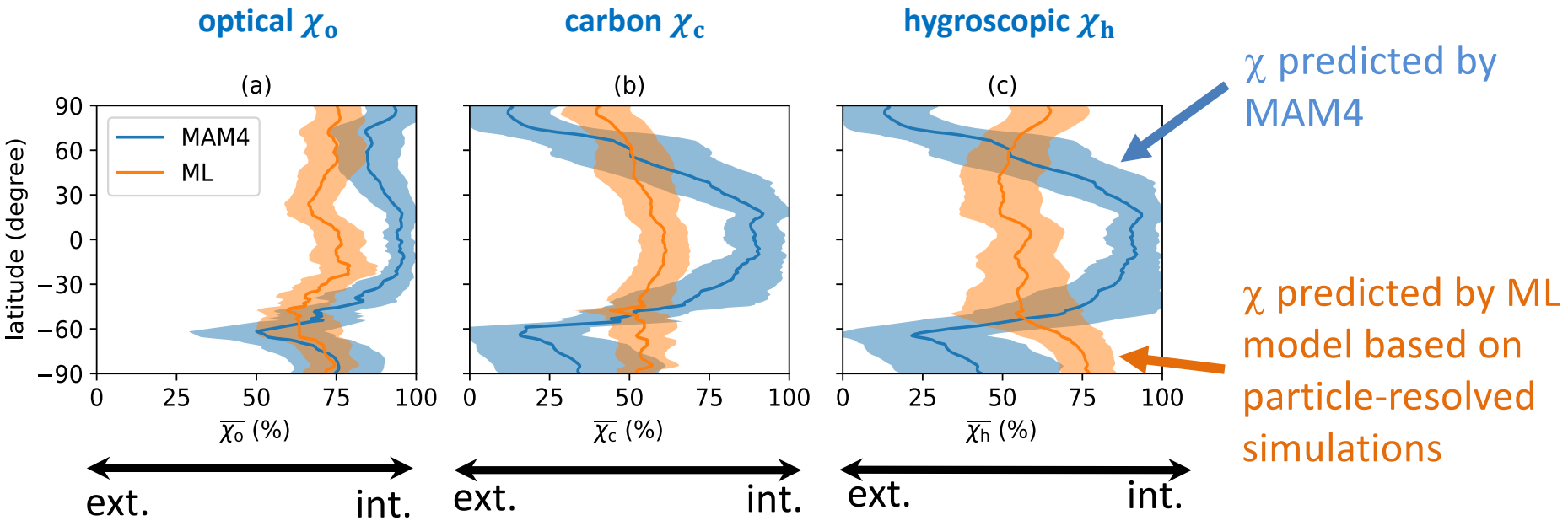
A. J. Sedlacek III, **E. R. Lewis**, T. B. Onasch, P. Zuidema, J. Redemann, D. Jaffe, and L. I. Kleinman, "Regime-dependent Mixing State of Black Carbon Particles Produced by Biomass Burn Events" *under review*

Mixing State: What are the questions?

- What is the aerosol mixing state of the ambient aerosol in different environments?

- How well can we represent mixing state in regional and global models?

Quantifying the structural uncertainty of the aerosol mixing state representation in MAM4



Objective

- Verify the global distribution of aerosol mixing state predicted by MAM4.

Key Finding

- MAM4 predictions of mixing state are too internally-mixed in low latitudes and too externally-mixed in high latitudes compared to the benchmark model.

Z. Zheng, M. West, L. Zhao, P.-L. Ma, X. Liu, N. Riemer, Quantifying the structural uncertainty of the aerosol mixing state representation in a modal model, Atmos. Chem. Phys., under review, 2021.

Aerosol direct impacts on climate: What are the questions?

- What do ARM measurements tell us about long-term trends in aerosol optical properties?

- How much absorption do black- and brown-carbon containing aerosol exert in different environments?

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Long-term Aerosol Trends and Radiative Effects over SGP

Objective

- Understanding the long-term aerosol changes and seasonality at SGP and impact on regional aerosol direct radiative effects (DRE)

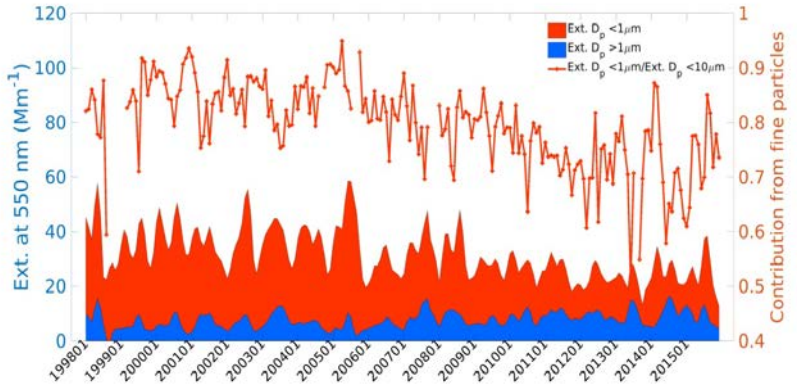
Method

- Analysis of the 18-year (1998-2015) ARM surface and column aerosol measurements and observationally-based radiative transfer calculations

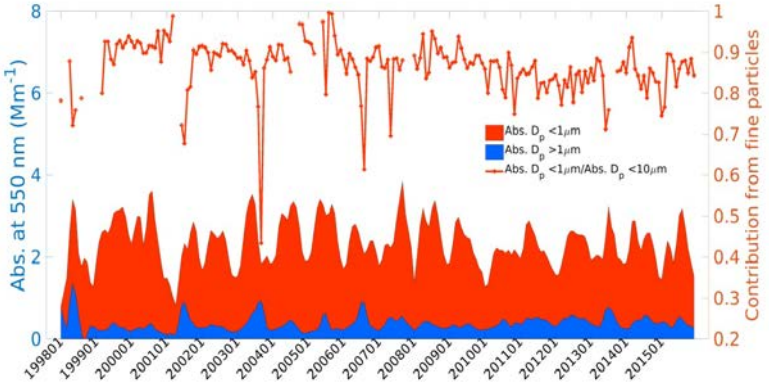
Key Findings

- AOD over SGP is found to decline at a slower rate (-1.04% yr^{-1}) than the more polluted regions in the continental US (~1.3% yr^{-1}).
- As Figure shows, the decrease of surface aerosol extinction is due to fine-mode aerosols, with a weak increase from coarse mode. Absorption is nearly constant during the last two decades.
- We also find a strong correlation (0.69) between surface extinction and AOD, suggesting column aerosol optical properties are strongly influenced by these surface aerosol changes.

Surface Aerosol Extinction



Surface Aerosol Absorption



As a result, the estimated clear-sky aerosol DRE at TOA decreases by -37% over the 18 years, with a smaller reduction in surface insolation (-27%). It provides the observationally based estimation of long-term aerosol radiative impact over the background continental US.

Y. Feng, L. Xu, A. McComiskey, and Art Sedlacek, "Long-term Trends and Seasonality of Column and Surface Aerosol Optical Properties over the Southern Great Plain", manuscript in prep. POSTER session 6, Thur 3-4pm

Aerosol direct impacts on climate: What are the questions?

- What do ARM measurements tell us about long-term trends in aerosol optical properties?

- How much absorption do black- and brown-carbon containing aerosol exert in different environments?

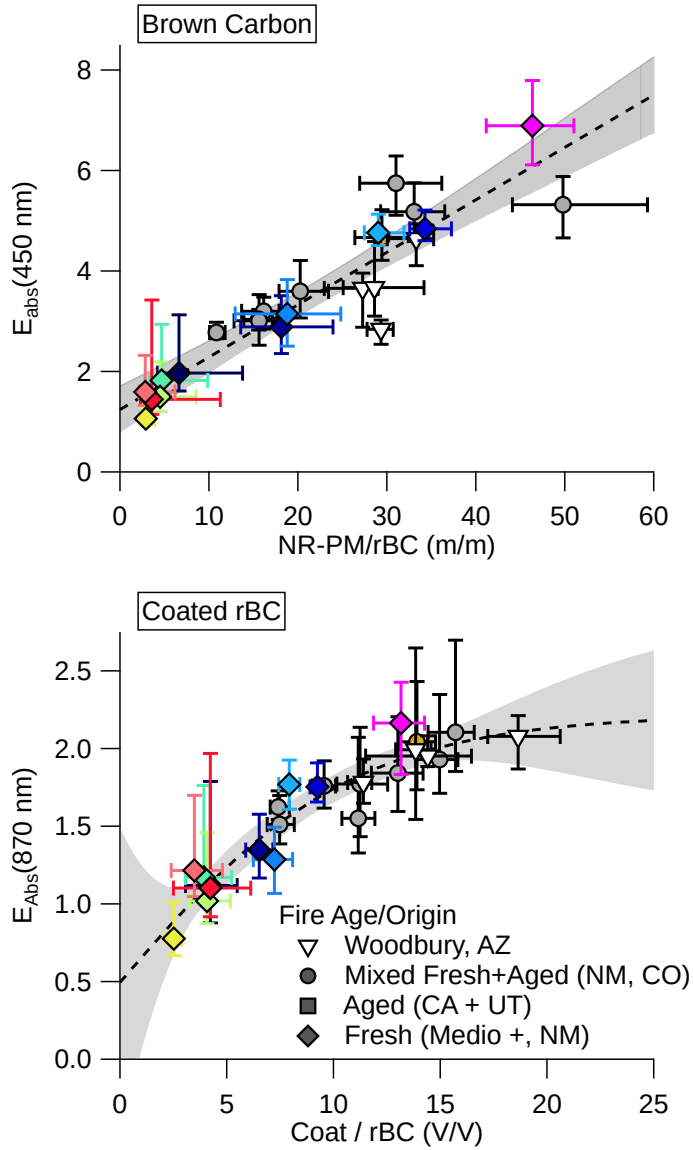
In situ observations of absorption enhancements in fresh-to-aged wildfire smoke plumes at Los Alamos, NM

Objective

- "Ground-truth" light-absorption enhancement models with observations of ambient wildfire smoke that span a wide range of fuel sources, combustion phases, atmospheric ages, and coating thicknesses.

Key Finding

- Absorption enhancements are driven by brown carbon (BrC) and coatings on black carbon (BC); two predictable processes that are well described by Mie approximations across all plume observations.
- Internally mixed BC in wildfire plumes can be treated as homogenously coated by organics.
- BrC of < ~1-day age has an effective MAC of ~1 m^2/g(Org), after which bleaching results in decreased absorptivity.



Ambient smoke absorption enhancements at blue (450 nm) and infrared (870 nm) wavelengths for 23 individual plumes of varying origin, fuels, and atmospheric age.

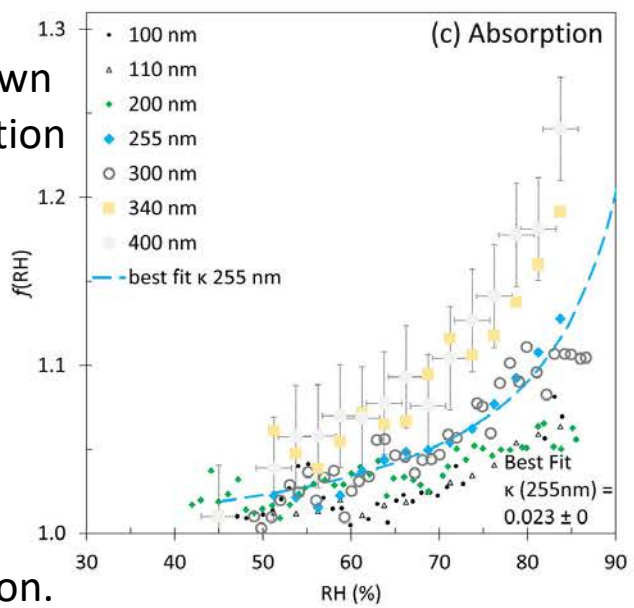
Lee, Gorkowski, Meyer, Benedict, Aiken & Dubey (LANL) in prep. PNAS '21

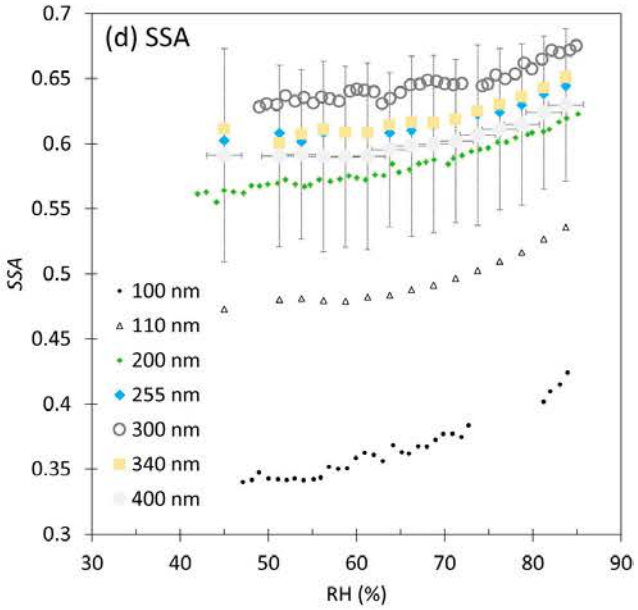
Observing RH-Dependent Absorbing Aerosol Optics: Gray and Brown Carbon (LANL)

Objective

- Humidity effects on black and brown carbon optics, particularly absorption are large in climate models but remain ill constrained due to measurement challenges.
- Develop a humidity (RH) control CAPS-PM_{SSA} to enable humidified aerosol observations reliably.
- Perform ambient and laboratory studies of RH dependent absorption.

f(RH) of Nigorsin dye (at 450 nm)

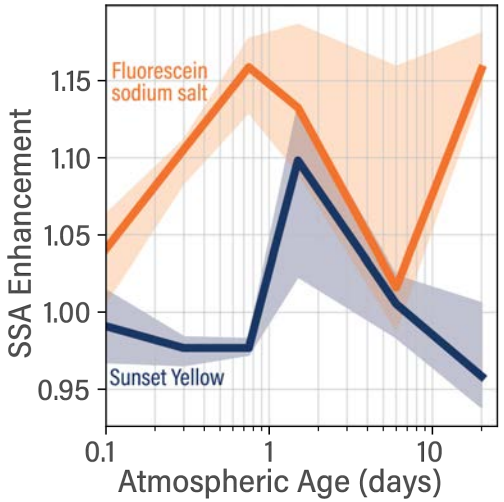




Key Finding

- Absorption enhancements can be large ~1.2 at 80% RH for nigrosin and depends on particle size.
- Absorption increase, that increases local heating, is smaller than the scattering and SSA increase, resulting in a net albedo brightening and TOA cooling.

Next steps are using a PAM reactor, to probe oxidation of brown carbon dyes and the influence on the humidity dependence.



•Carrico, C. M., Capek, T. J., Gorkowski, K. J., Lam, J. T., Gulick, S., Karacaoglu, J., Lee, J. E., Dungan, C., Aiken, A. C., Onasch, T. B., Freedman, A., Mazzoleni, C., & Dubey, M. K. (2021). Humidified single-scattering albedometer (H-CAPS-PM SSA): Design, data analysis, and validation. *Aerosol Science and Technology*, 1–20. https://doi.org/10.1080/02786826.2021.1895430

Aerosol indirect impacts on climate: What are the questions?

- How do clouds modify aerosols?
- How well can we predict the concentrations of cloud condensation nuclei and of ice nucleating particles if we know aerosol size and composition?
- How effective are different particle types in forming ice? (and what does this mean for ice cloud properties?)

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Soot Compaction Upon Cloud Processing

Objective

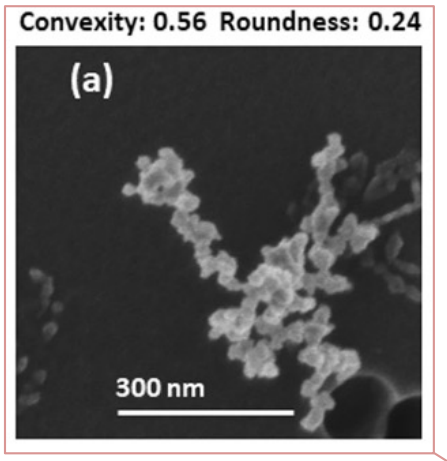
- Understand the effects of cloud processing on soot particles

Key Finding

- Soot particles after evaporating cloud droplets are more compact (more convex) than freshly emitted and interstitial soot

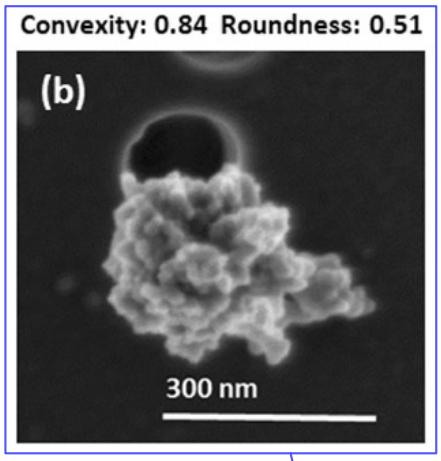
Implications

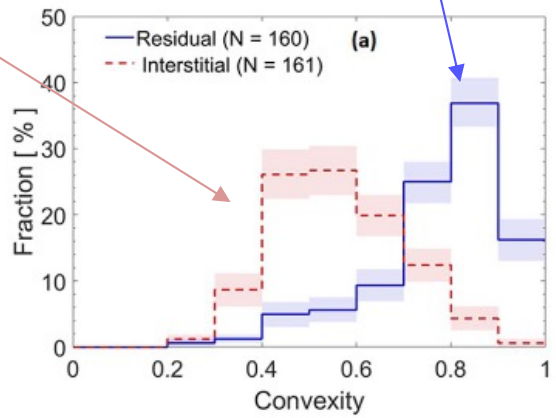
- Compaction can affect the fate, radiative, surface, aerodynamic, and ice nucleating properties of soot particles



Cloud formation

Soot aggregate compaction





Soot compaction upon cloud formation in the MTU PI-Chamber

Bhandari, J., S. China, K. K. Chandrakar, G. Kinney, W. Cantrell, R. A. Shaw, L. R. Mazzoleni, G. Girotto, N. Sharma, K. Gorkowski, S. Gilardoni, S. Decesari, M. C. Facchini, N. Zanca, G. Pavese, F. Esposito, M. K. Dubey, A. C. Aiken, R. K. Chakrabarty, H. Moosmüller, T. B. Onasch, R. A. Zaveri, B. V. Scarnato, P. Fialho and C. Mazzoleni (2019). "Extensive Soot Compaction by Cloud Processing from Laboratory and Field Observations." Scientific Reports 9(1): 11824.

Aerosol indirect impacts on climate: What are the questions?

- How do clouds modify aerosols?

- How well can we predict the concentrations of cloud condensation nuclei and of ice nucleating particles if we know aerosol size and composition?

- How effective are different particle types in forming ice?

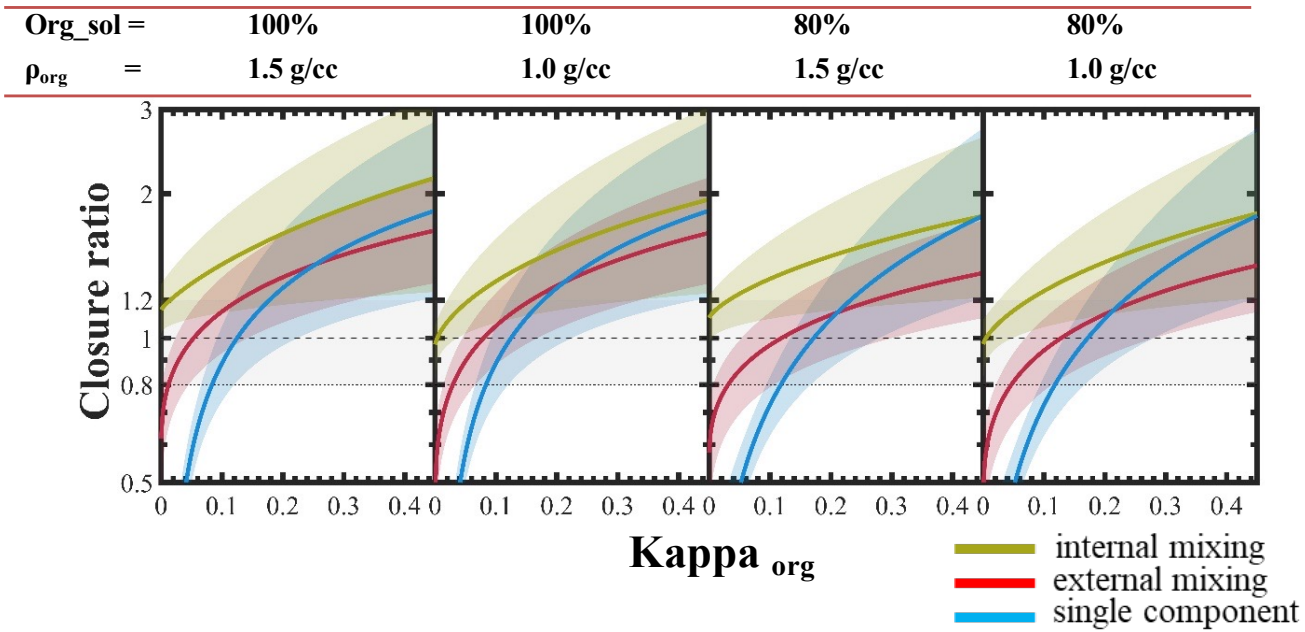
Airborne CCN closure study using HI-SCALE measurements

Objective

- Assess the importance of composition and aerosol mixing state to better represent CCN concentration in large-scale model.

Key Finding

- We find that CCN prediction is sensitive to the assumption of aerosol mixing state and organic kappa values, but not so much to the organic density and solubility.



CCN closure was assessed using different permutations of organic aerosol density, kappa, and solubility factors for each three size-independent mixing state assumptions at two supersaturations and seasons.

Kulkarni, G., Mei, F., Shilling, J., Wang, J., Flynn, C., Zelenyuk-Imre, A. and Fast, J.: "Factors affecting the CCN closure at the SGP site based on airborne HI-SCALE campaign measurements" – under preparation

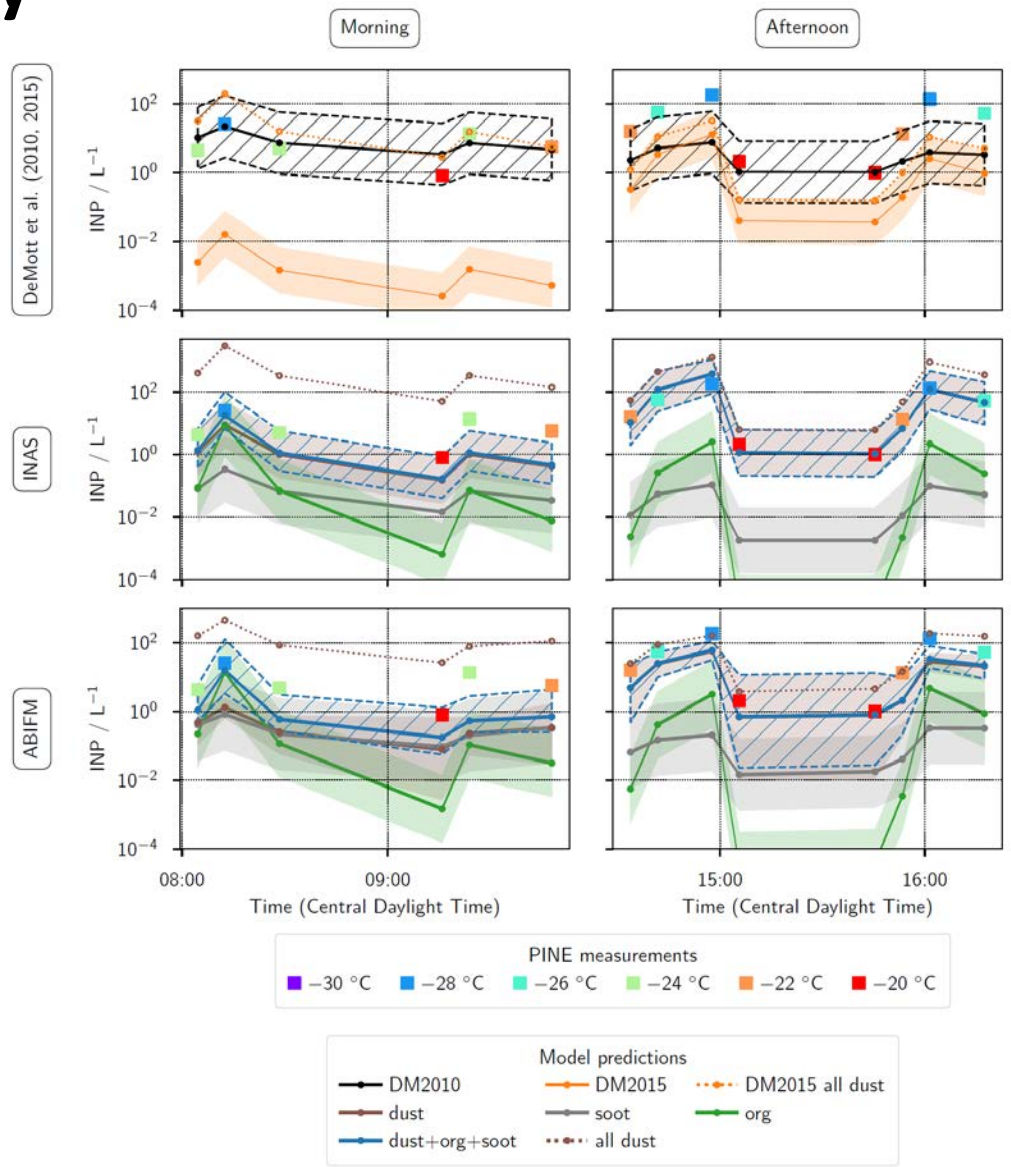
AEROICESTUDY: An ARM Southern Great Plains Aerosol-Ice Formation Closure Pilot Study

Objectives

- *Identify robust ice nucleation parameterizations.*
- *Determine the crucial aerosol physicochemical properties to guide models and long-term INP measurements.*
- *Assess the level of parameter details to achieve aerosol-INP closure.*
- *What are the leading causes for climate model bias in INP predictions.*

Key Findings

- *Assessed experimental prerequisites to achieve aerosol-INP closure.*
- *Partial and full closure was achieved by different parameterizations and was dependent on knowledge of ambient INP types.*
- *INP characterization of inorganic-organic, soil-organic, and biological particles is needed to improve closure.*
- *Physicochemical characterization of ambient aerosol population is crucial.*



Knopf, D. A., Barry, K. R., Brubaker, T. A., Jahl, L. G., Jankowski, K. A., Li, J., Lu, Y., Monroe, L. W., Moore, K. A., Rivera-Adorno, F. A., Sauceda, K. A., Shi, Y., Tomlin, J. M., Vepuri, H. S. K., Wang, P., Lata, N. N., Levin, E. J. T., Creamean, J. M., Hill, T. C. J., China, S., Alpert, P. A., Moffet, R. C., Hiranuma, N., Sullivan, R. C., Fridlind, A. M., West, M., Riemer, N., Laskin, A., DeMott, P. J., Liu, X., *Bulletin of the American Meteorological Society*, 2021, *accepted*.

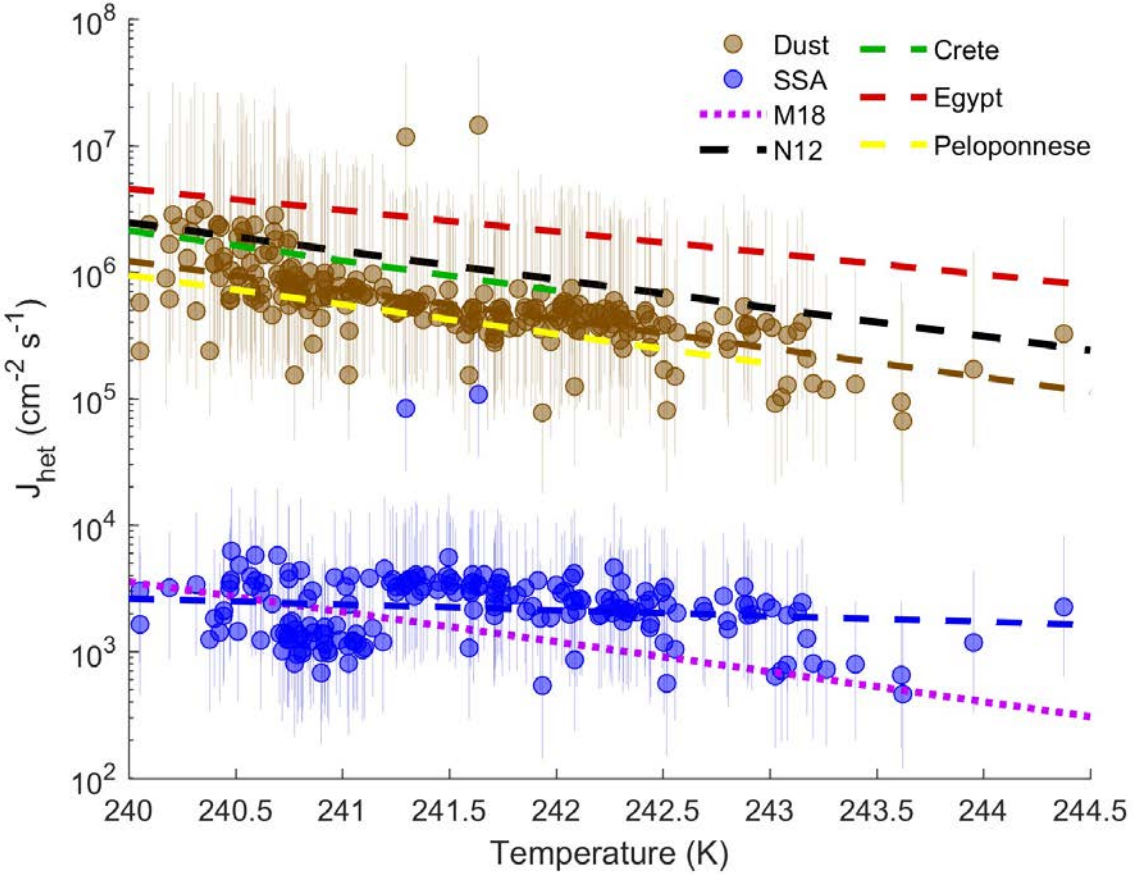
A novel method for determining heterogeneous freezing rates (J_{het}) for multiple classes of ambient particles

Objective

- Calculate freezing rates (J_{het}) for ambient dust and sea spray aerosol under field conditions.
- Compare with previous parameterizations of freezing by dust and sea spray INPs.

Key Finding

- We introduce a novel method for direct calculation of immersion freezing rates for samples of ambient particles, resolved by particle type (dust versus sea spray).
- Single particle mass spectrometry enables J_{het} calculation for multiple particle types simultaneously.
- Particles sample sizes are largest source of uncertainty, mitigated by use of aerosol concentrator.



Immersion freezing rates (J_{het}) as a function of temperature, calculated for dust and sea spray aerosol characterized with single-particle mass spectrometry during field measurements at Bodega Bay. Field measurements were collected in parallel with the ACAPEX campaign.

G. C. Cornwell, C. S. McCluskey, P. J. DeMott, K. A. Prather, S. M. Burrows, "Determining heterogeneous ice nucleation rate coefficients from ambient measurements", *in prep.*

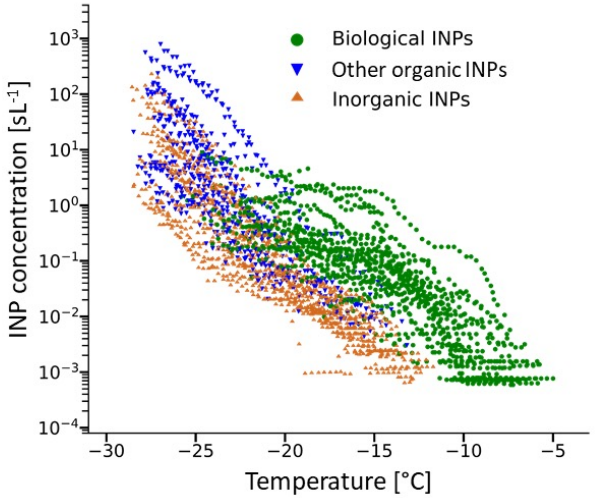
Aerosol indirect impacts on climate: What are the questions?

- How do clouds modify aerosols?
- How well can we predict the concentrations of cloud condensation nuclei and of ice nucleating particles if we know aerosol size and composition?
- How effective are different particle types in forming ice? (and what does this mean for ice cloud properties?)

Ice nucleating particle (INP) typing defines their relation to land sources and meteorology

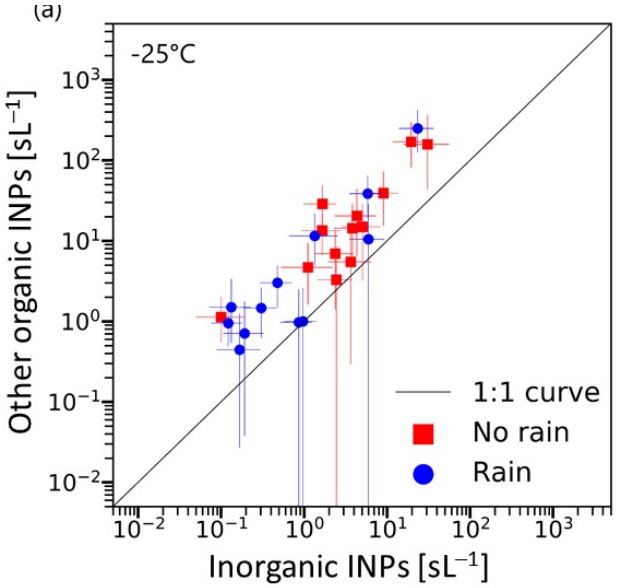
Objective

- Understand the factors driving the makeup and temporal variability of INPs over the Cloud, Aerosol, and Complex Terrain Interactions (CACTI) region using immersion freezing methods, combined with ARM aerosol and meteorological data.
- INP types defined by thermal and chemical neutralization of biological and organic components. Basis for parameterizations.



Key Findings

- Biological (at higher T) and other organic (at lower T) INPs dominate, the former driven positively by rain events, and vice versa. Inorganic INPs contribute least.
- Relationship between organic and inorganic INPs, combined with air mass analysis, defines constant regional arable soil source.
- Comparison to laboratory INP and mass spectral measurements of soil particles confirms source, and felsic/illitic mineral influences inferred from active site density analysis.



Acknowledgment: DE- SC0021116

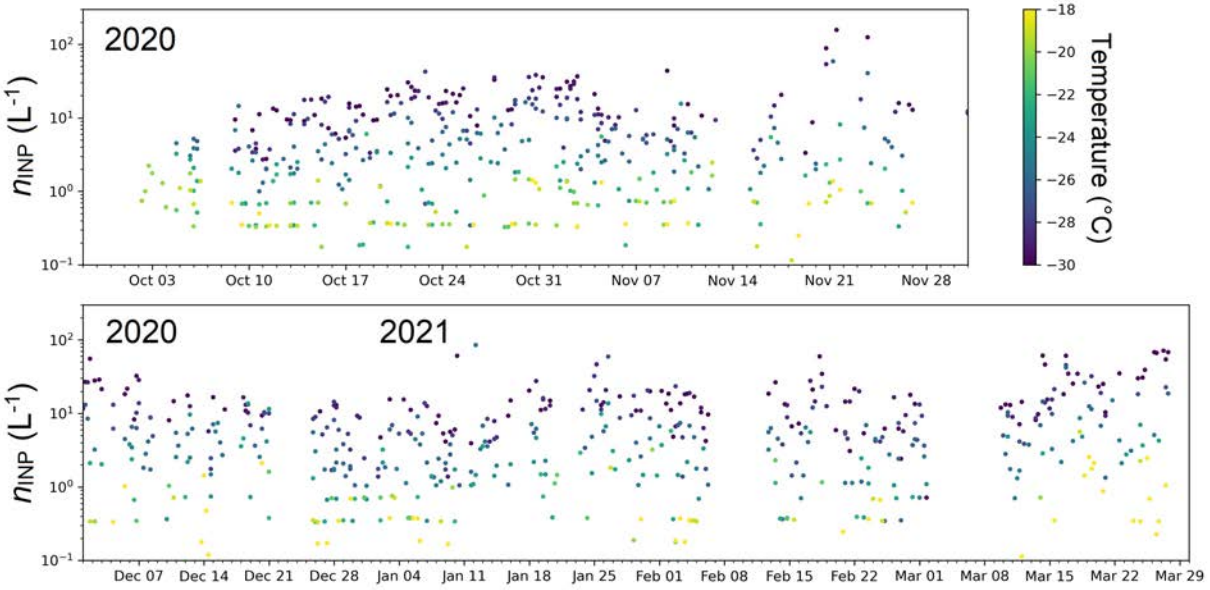
Testa, B., T. C. J. Hill, N. Marsden, K. R. Barry, C. C. Hume, Q. Bian, J. Uetake, H. Hare, R. J. Perkins, O. Möhler, S. M. Kreidenweis, and P. J. DeMott, 2021: Ice nucleating particles in the boundary layer of the Sierras de Córdoba, Argentina, during the Cloud, Aerosol, and Complex Terrain Interactions experiment, in review to J. Geophys. Res., preprint available in Earth and Space Science Open Archive, https://doi.org/10.1002/essoar.10506960.1

Ice-nucleating particle concentration measurements from the ARM Eastern North Atlantic (ENA) and Southern Great Plains (SGP) sites

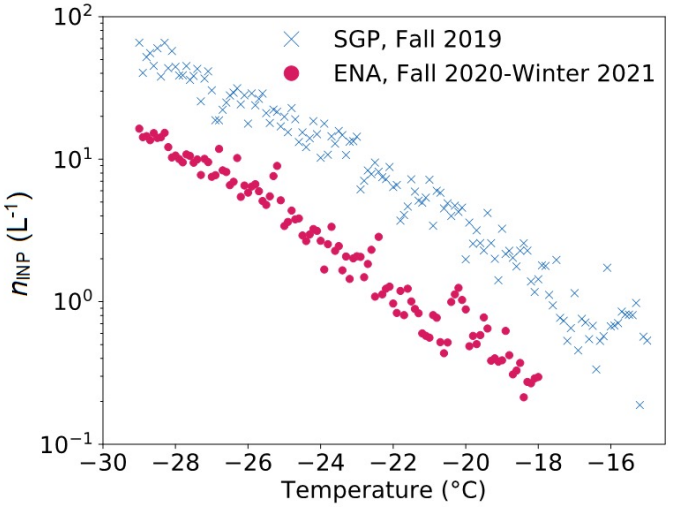
Objective: Generating new ambient INP data from the ARM mega-sites

PI: Naruki Hiranuma
nhiranuma@wtamu.edu

Key Findings: The terrestrial SGP site has higher nINP across the range of examined temperatures than nINP from the marine predominant ENA[1]. A moderate correlation between nINP and cloud condensation nuclei concentration (-20 °C, 0.1% super saturation) was observed at ENA ($\rho = 0.47$, $p<0.001$), and the correlation was higher in Fall ($\rho = 0.81$).



The time series plot of the ice crystal concentration (n_{ice}, L^{-1} air) for temperatures below -18 °C during the ENA campaign. The portable ice nucleation experiment chamber[2] measured n_{ice} for ~200 days from October 2020 to March 2021. Time averaged data (6 hour) of n_{ice} at the end of individual expansion are shown.



Temperature-binned average n_{ice} spectra during SGP campaign (blue Xs) and ENA campaign (pink dots) at each 0.1 °C. The data uncertainties are discussed and presented in the PI's poster.

1) Wilbourn, E. K. et al.: Remote ice-nucleating particle measurements from the Eastern North Atlantic during autumn and winter, https://doi.org/10.5194/egusphere-egu21-6314, EGU General Assembly, Online, April 27, 2021.
2) Möhler, O. et al.: The Portable Ice Nucleation Experiment (PINE): a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles, Atmos. Meas. Tech., 14, 1143–1166, https://doi.org/10.5194/amt-14-1143-2021, 2021.

Impact of marine organic ice-nucleating particles on high-latitude mixed-phase clouds

Objective

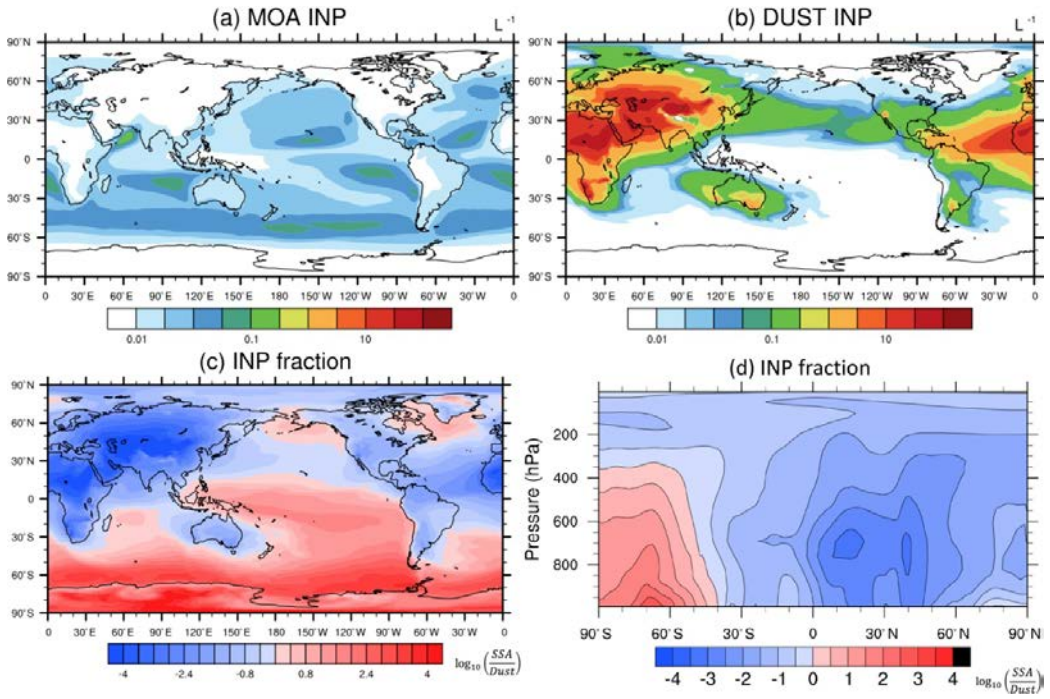
- Study the contribution of marine organic aerosols (MOA) through immersion mode to ice-nucleating particle (INP) concentrations and impacts on mixed-phase clouds in high latitudes.

Approach

- Implement MOA as a new aerosol species into a global climate model, and enable MOA aerosol–cloud interactions via aerosol activation and ice nucleation;
- Compare modeled versus measured INPs from different campaigns**;** Study the impact of MOA INPs on mixed-phase clouds.

Impact

- MOA INPs through immersion mode dominate ice nucleation below 400 hPa over the Southern Ocean, while dust INPs are more abundant elsewhere.
- Including the contribution of MOA INPs improves the model agreement with INP observations over high latitudes.
- By acting as INPs, MOA enhances the longwave cloud forcing by 0.35 W m^{-2} in the austral winter over the Southern Hemisphere (20-90 S).



Spatial distribution of annual mean concentrations of (a) MOA INPs, (b) dust INPs, and (c) ratio of MOA INP concentration to dust INP concentration at 950 hPa, and (d) vertical cross sections of ratio of MOA INP concentration to dust INP concentration. INP concentrations are diagnosed at temperature of –25°C using McCluskey et al. (2018) and DeMott et al. (2015) for dust and MOA, respectively, with MOA and dust concentrations predicted from CESM2.

Zhao, X., Liu, X., Burrows, S. M. and Shi, Y.: Effects of marine organic aerosols as sources of immersion-mode ice-nucleating particles on high-latitude mixed-phase clouds, Atmos. Chem. Phys., 21(4), 2305–2327, doi:10.5194/acp-21-2305-2021, 2021.

A new ARM initiative: the measurement of ice nucleating particles (INPs)

- INP mentors: Jessie Creamean and Tom Hill
- Started in 2020 for INP concentrations from 24-h filters collected ~ every 6 days at ARM sites
- FY21 locations included SGP and AMF3
- FY22 locations will include SGP, TRACER, and SAIL
- Data will be available on the ARM Data Archive starting summer 2021
  - Total aerosol INP concentrations for all samples
  - Heat-labile and organic INPs on 1/3 of all samples
- Duplicate filters preserved for users
- PI requests can be made for INP measurements!

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