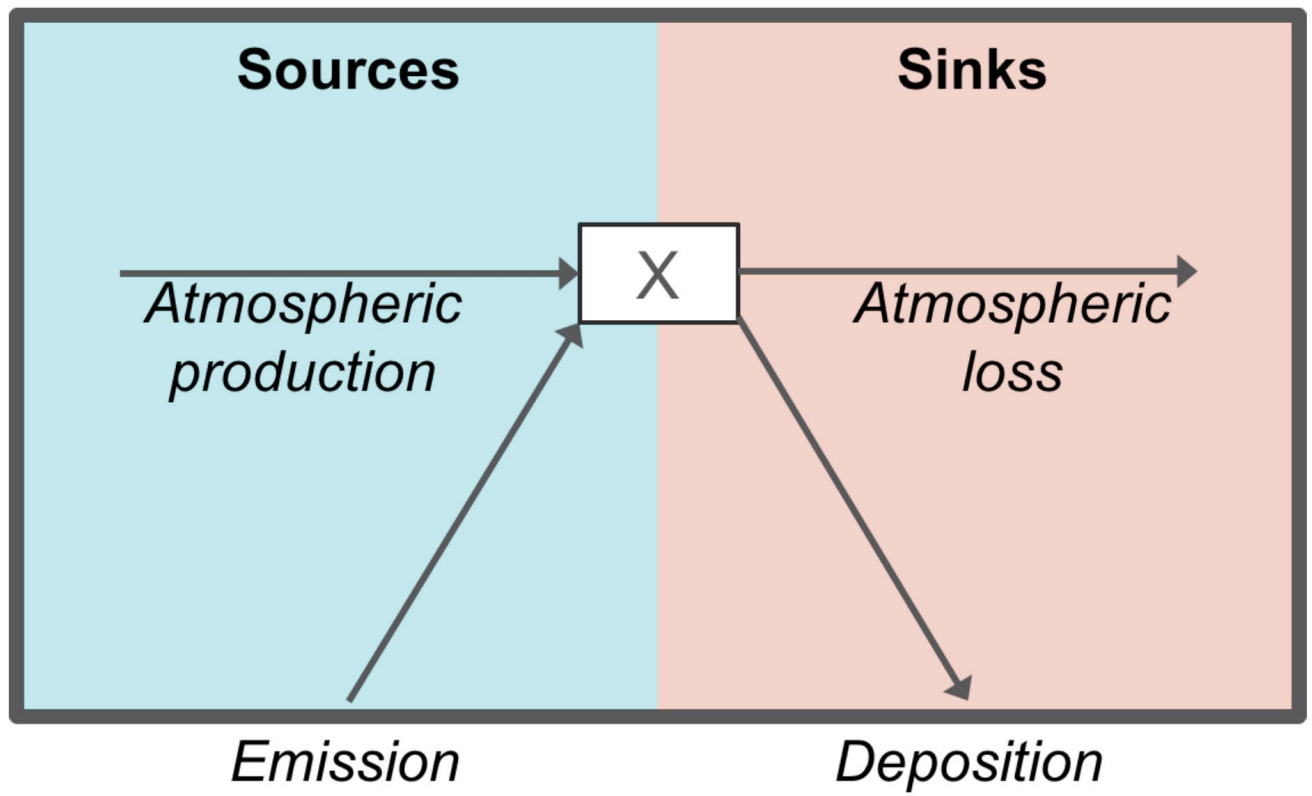
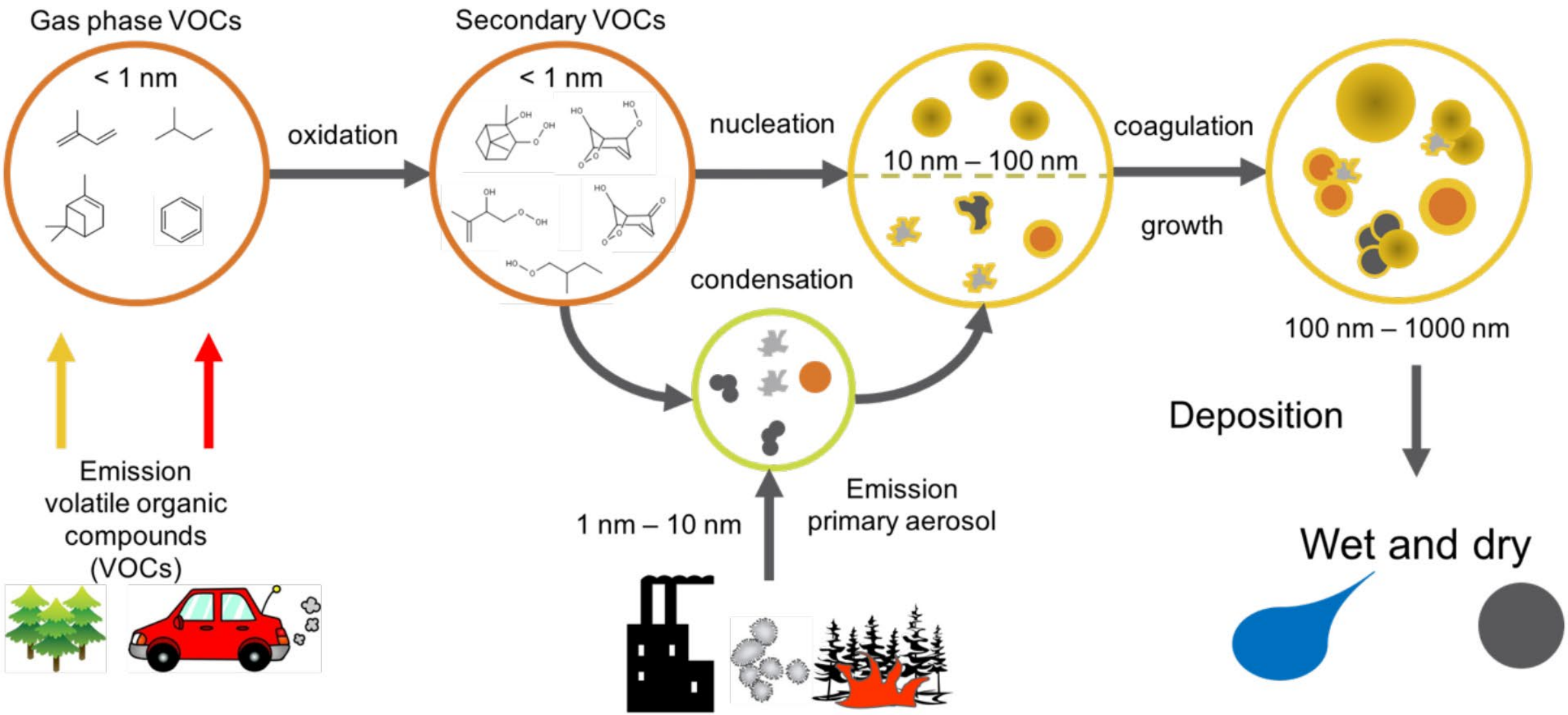
Sources and fate of submicron particles:
How observations can constrain emissions and deposition

Delphine K. Farmer, Lauren Garofalo, Matson Pothier,
Sonia Kreidenweis, Ezra Levin, Ethan Emerson
Colorado State University


Sources
Sinks
X
Atmospheric production
Atmospheric loss
Emission
Deposition


Gas phase VOCs
< 1 nm
oxidation
Secondary VOCs
< 1 nm
nucleation
10 nm – 100 nm
coagulation
growth
100 nm – 1000 nm
condensation
Deposition
Emission
volatile organic
compounds
(VOCs)
1 nm – 10 nm
Emission
primary aerosol
Wet and dry

AQUARIUS-relevant aerosol questions

- How do organic and inorganic components contribute to wintertime PM?
  Organics NHx, NOx, Cl
- What is the size-dependence of composition?
- What chemistry is controlling PM mass and composition, and how does this vary with meteorology?
- How accurate are emissions inventories (traffic, agriculture, VCPs, etc)?
- How does deposition affect gas and particle fate – and thus chemistry?

Observational constraints on atmospheric chemical processes

Ground sites

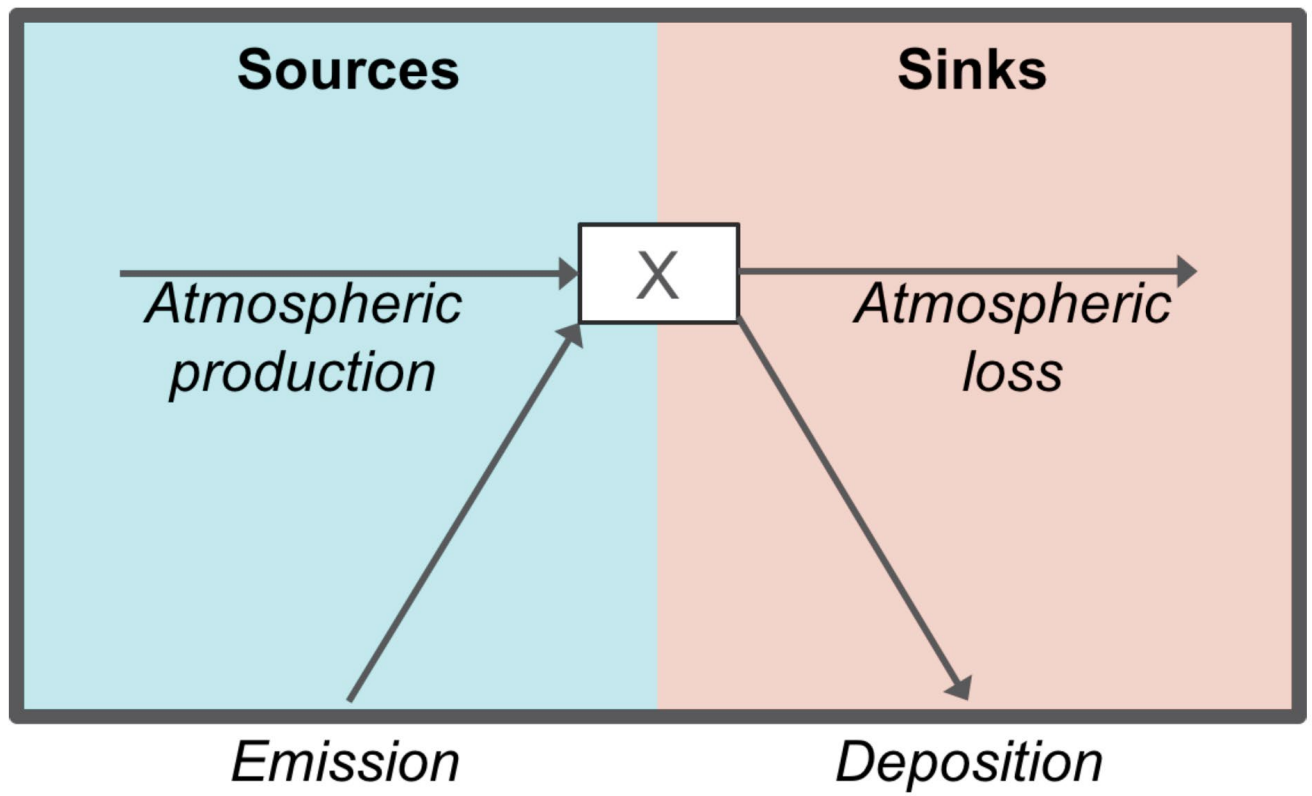
- Detailed chemical analysis
- Longer-term measurements
- Temporal variability
- Flux measurements: emission & deposition
- Single point locations

Example of ground-based flux measurements to think about particle dry deposition

Aircraft platform

- Chemical processes, emissions
- (Slightly less) detailed chemical analysis
- Fast time-resolution measurements
- Spatial & vertical gradients
- Multiple locations

Example of using bulk aerosol measurements to study emissions & chemistry


Sources
Sinks
X
Atmospheric production
Atmospheric loss
Emission
Deposition

Investigating dry deposition by size-resolved particle & black carbon flux measurements (eddy covariance + UHSAS, SP2)

Manitou Forest, CO (NOAA)

4 seasons, 2015

*array of turbulence conditions

Southern Great Plains, OK (DOE)

6 weeks, 2016

Investigating dry deposition by size-resolved particle & black carbon flux measurements (eddy covariance + UHSAS, SP2)

Manitou Forest, CO (NOAA)

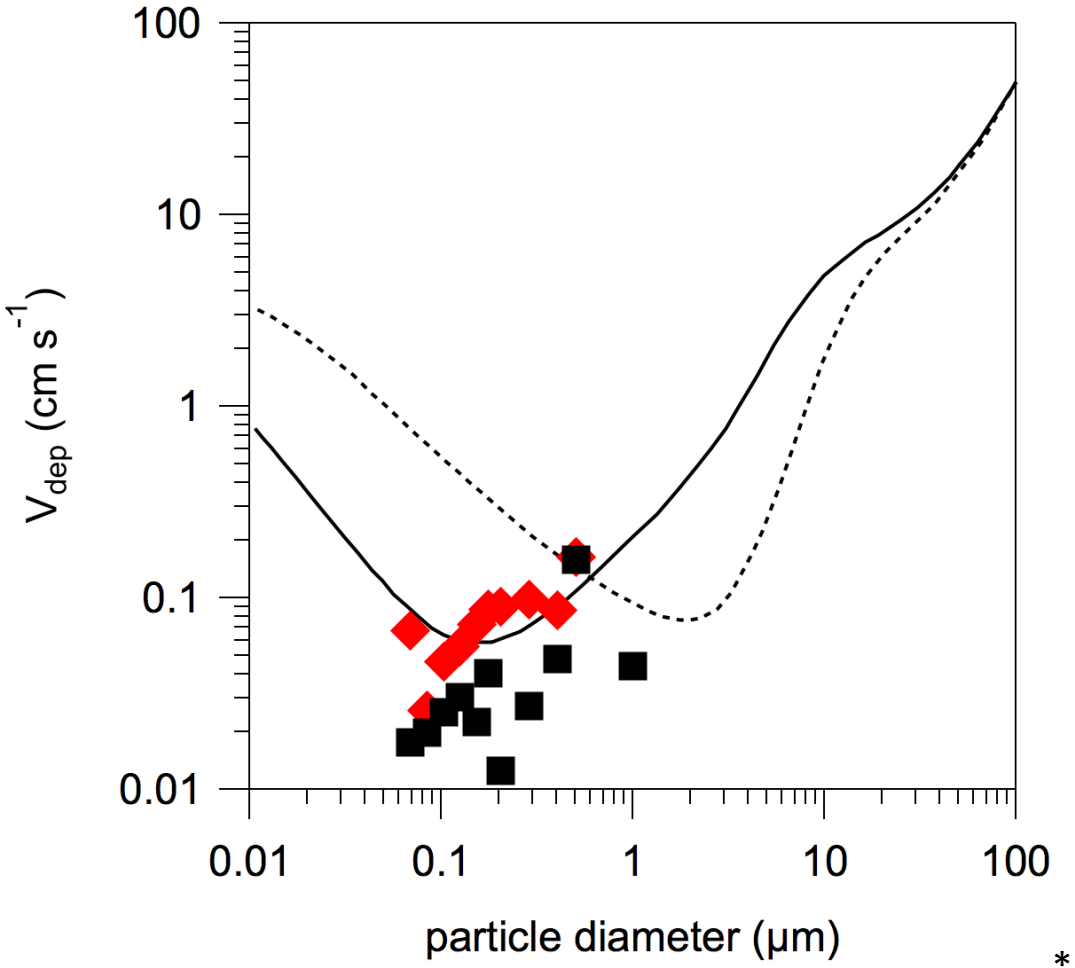
4 seasons, 2015

*array of turbulence conditions

Southern Great Plains, OK (DOE)

6 weeks, 2016

Sophisticated deposition models capture the observations (but widely used simpler ones generally do not)

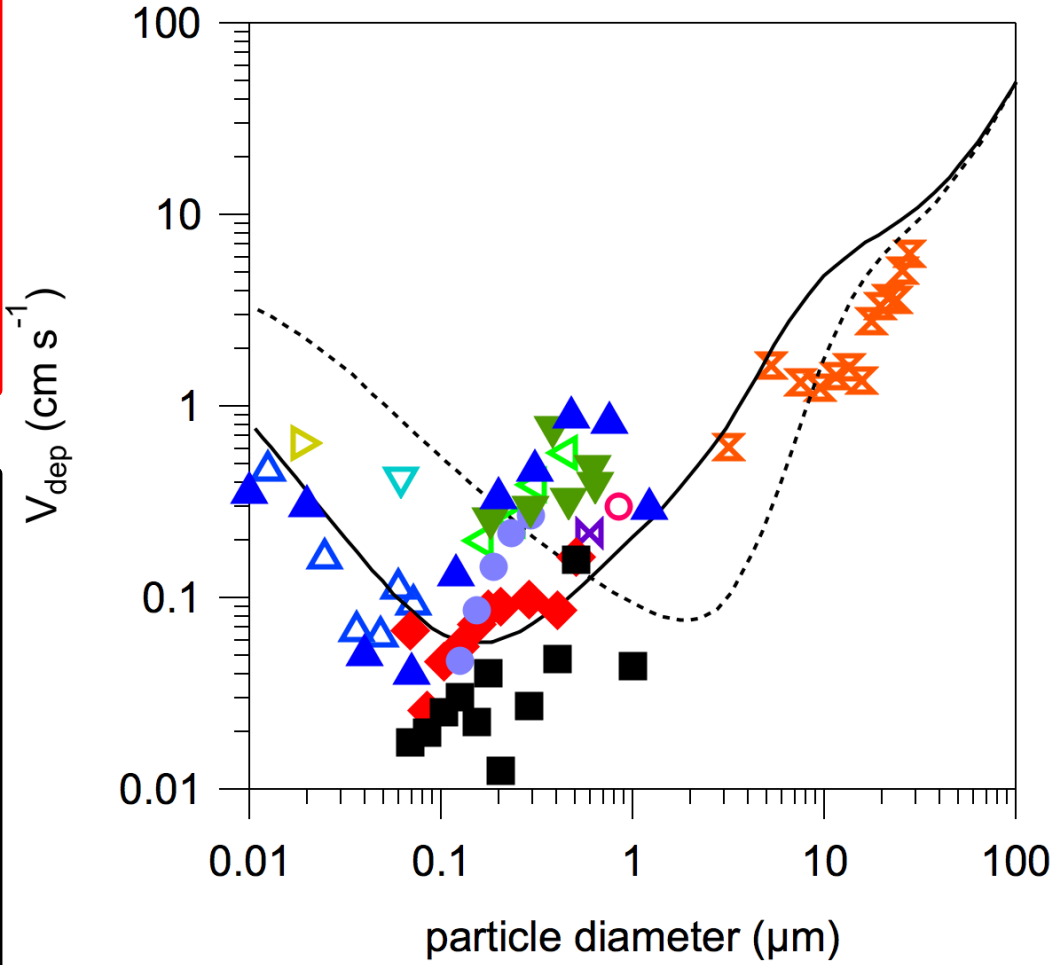


Zhang 2001 (model)
Petroff 2010 (model)
open symbols considered in Petroff 2010 model update
our data (pine forest)
our data (plains)

* Model shown is for forested terrain

Zhang et al., (2001); Petroff & Zhang, (2010)

Sophisticated deposition models capture the observations (but widely used simpler ones generally do not)

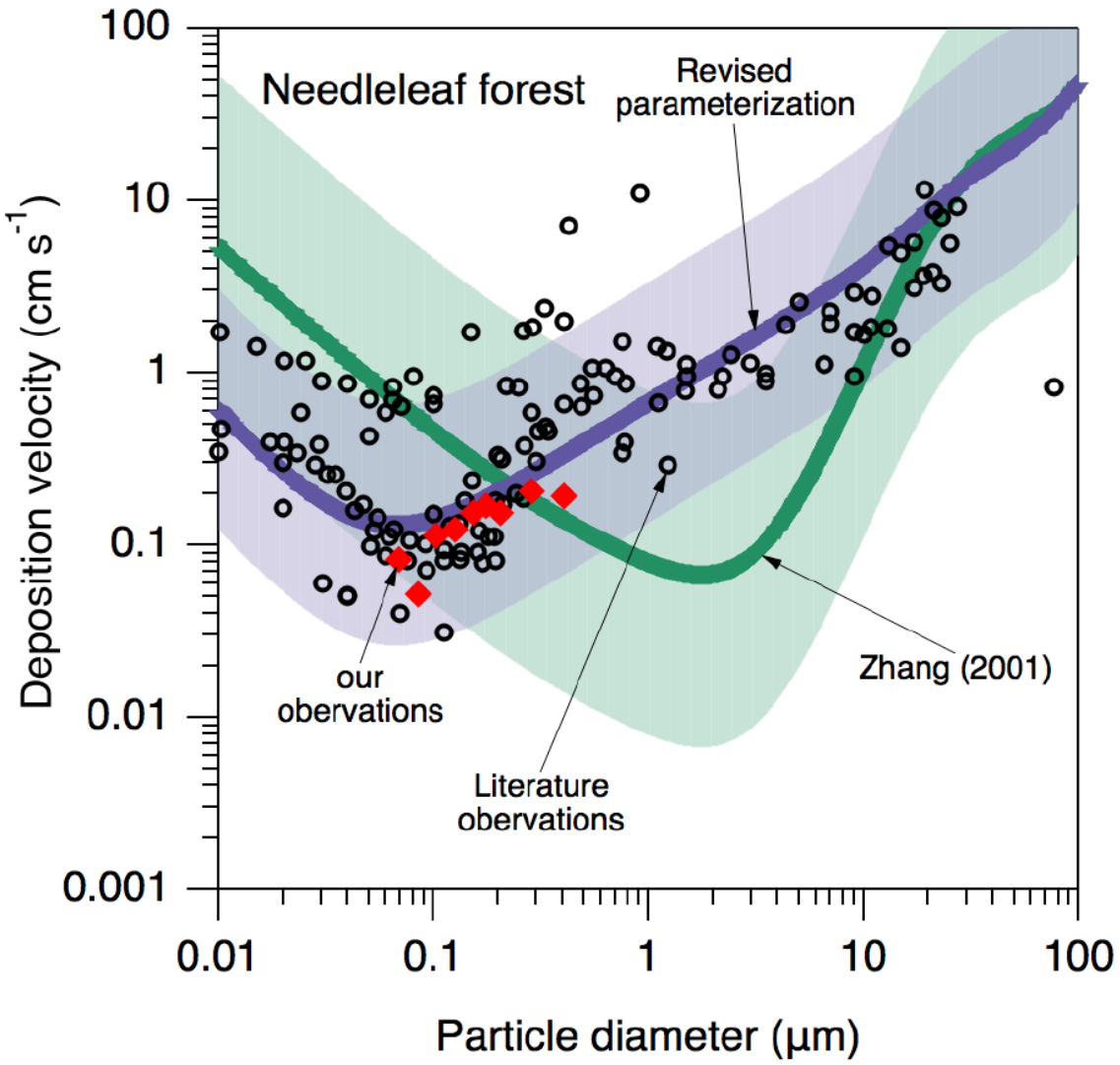


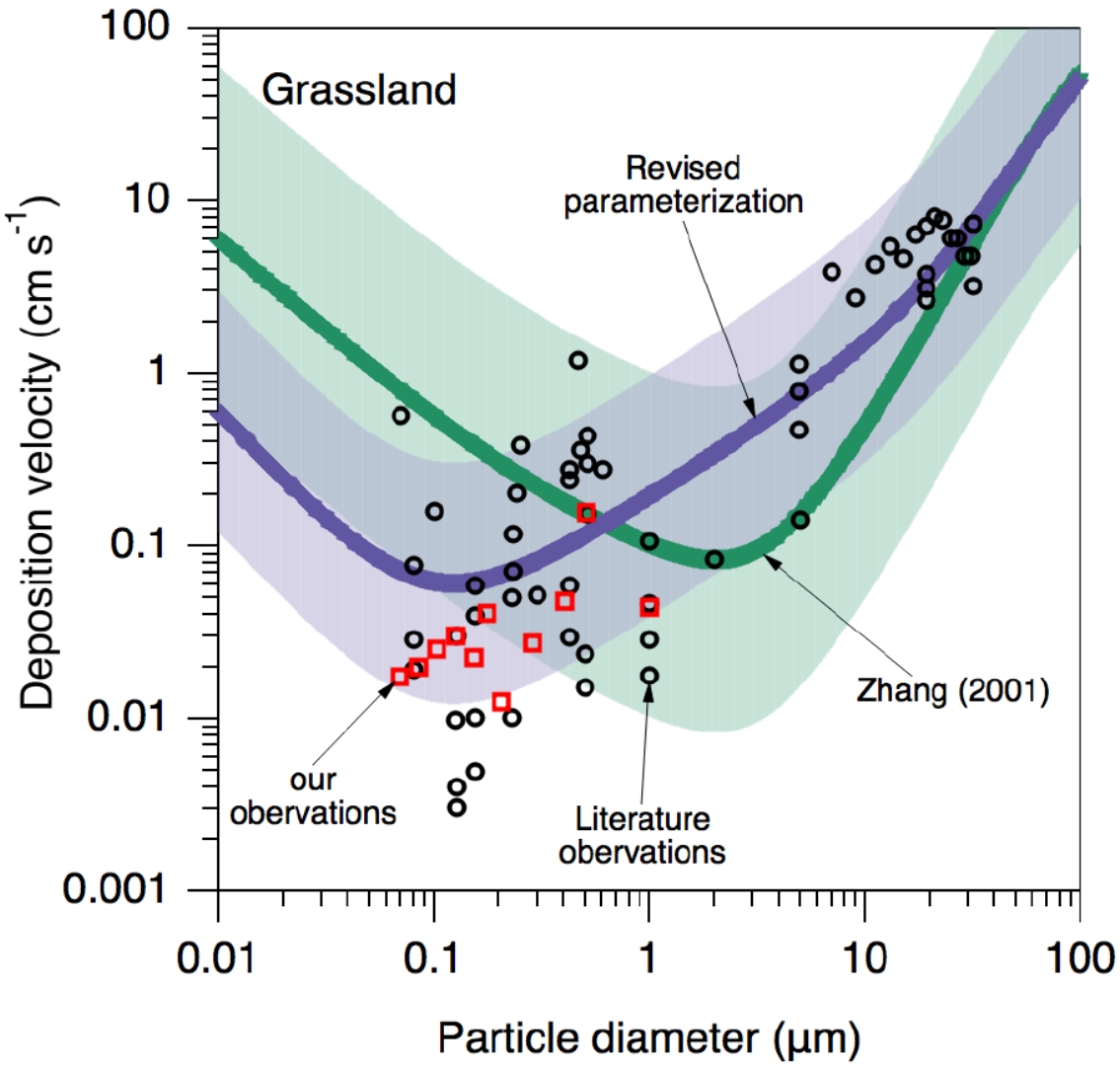
Zhang 2001 (model)
Petroff 2010 (model)
open symbols considered in Petroff 2010 model update
Lorenz 1989 (pine)
Beswick 1991 (spruce)
Lamaud 1994 (pine)
Gallager 1997 (fir)
Buzorius 2000 (pine)
Gaman 2004 (pine)
Vong 2007 (pine)
Gronholm 2009 (pine)
Deventer 2015 (spruce)
Petroff 2018 (broadleaf)
our data (pine forest)
our data (plains)

* Model shown is for forested terrain

Zhang et al., (2001); Petroff & Zhang, (2010)

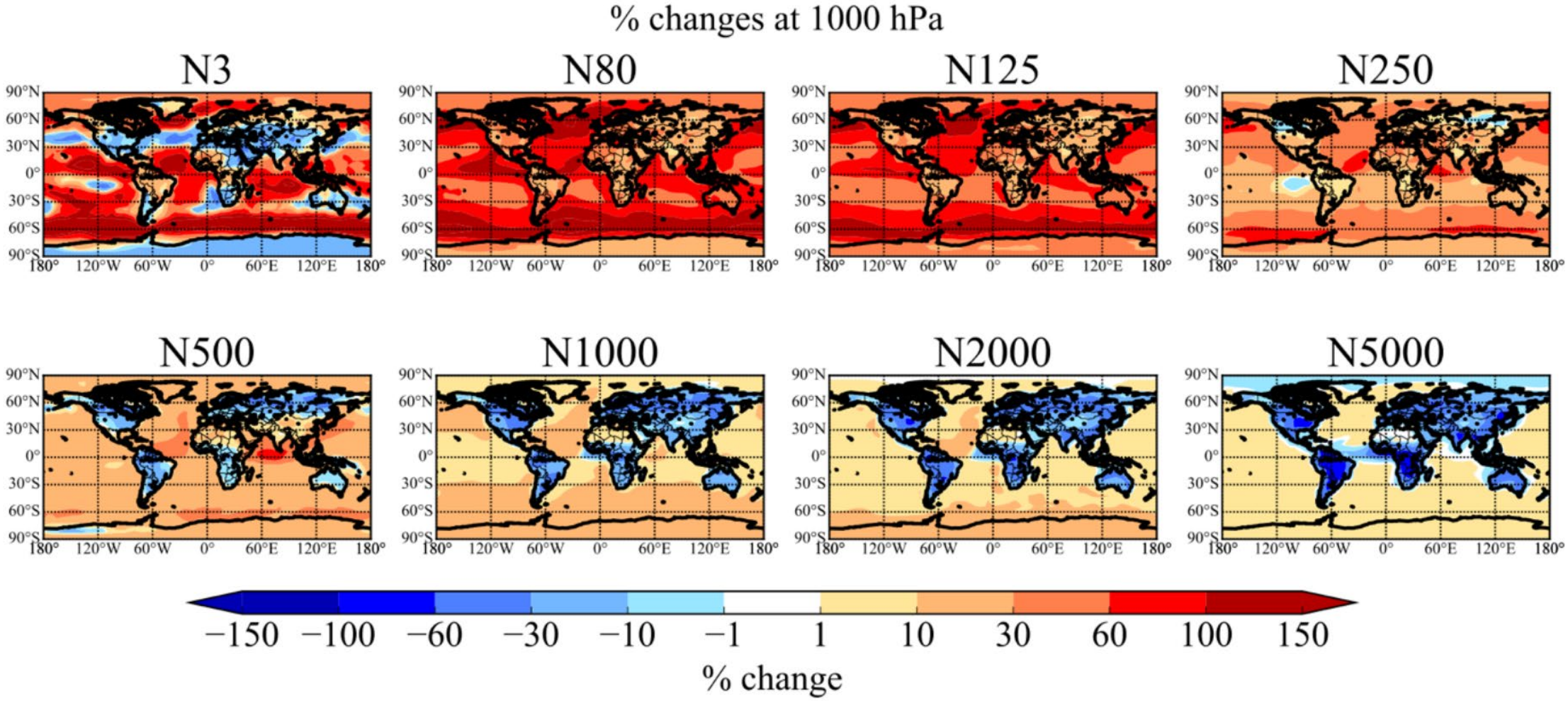
Standard vs revised parameterizations for dry deposition impact (size-resolved) particle lifetime





[Emerson et al. In prep]

Revised particle dry deposition parameterizations have a substantial effect on modeled aerosols + radiative effects



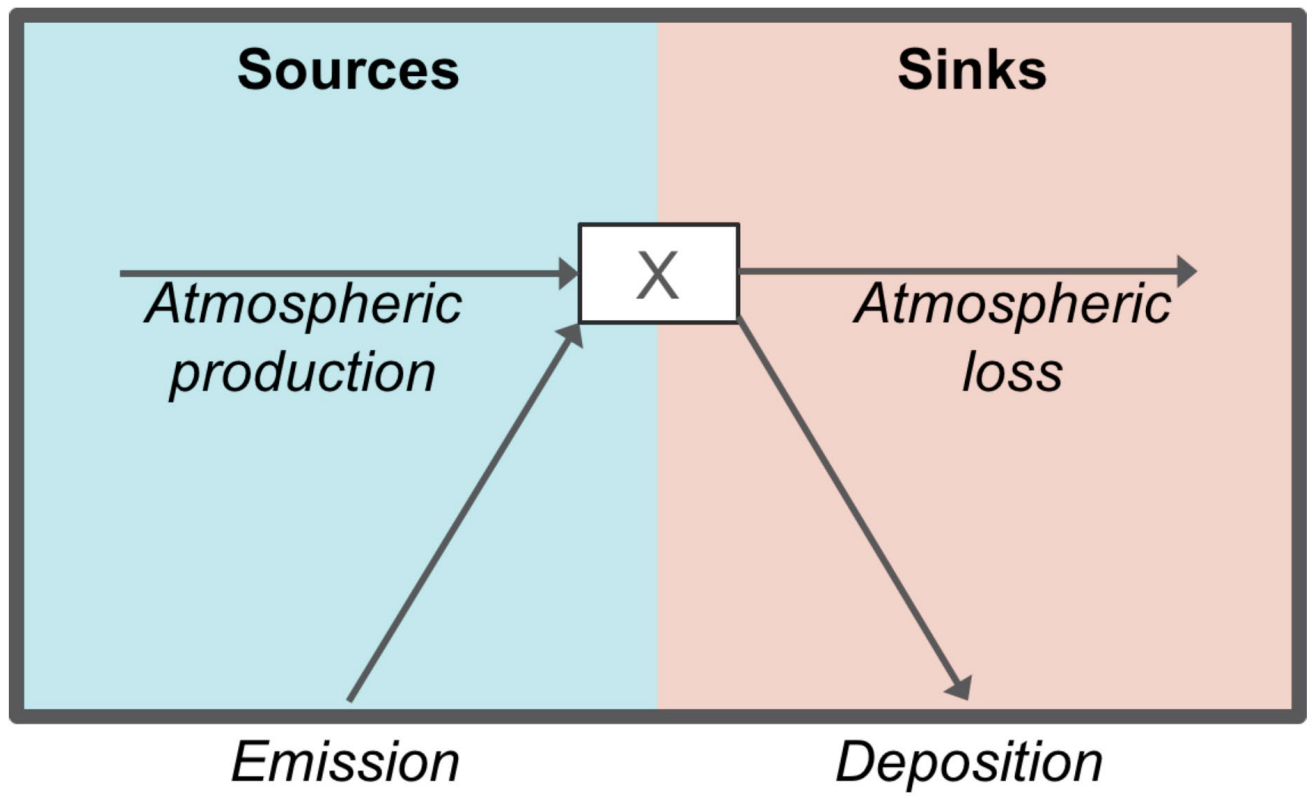
*Collaboration with Anna Hodshire + Jeff Pierce (CSU); Manuscript in prep

- We can use flux observations to constrain model parameterizations of sources & sinks

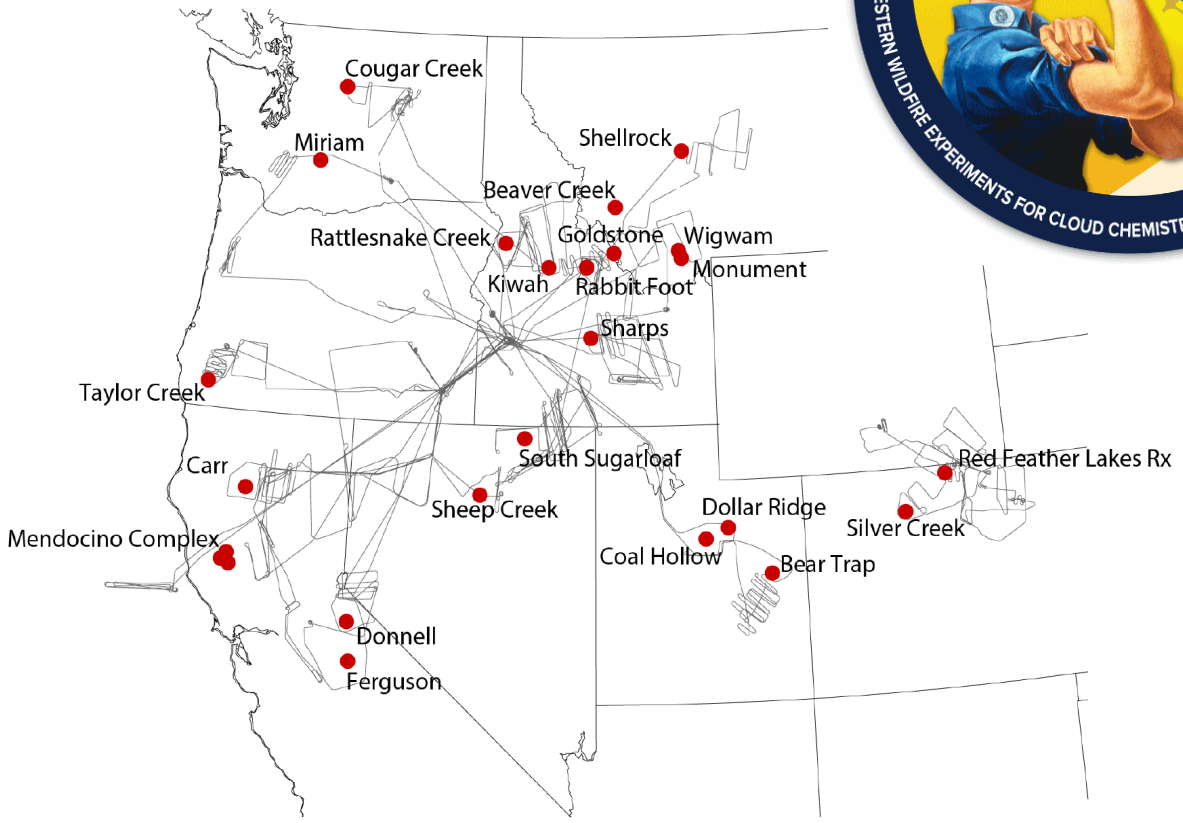
- Particularly large particle deposition uncertainties over cryosphere & water

- Remote / receptor sites offer an opportunity to investigate particle fate – and other impacts of deposition on C,N cycles – using flux measurements

- In contrast, urban & agricultural sites offer opportunity to study source emissions by direct flux measurements (VCPs, BC, NH_3)
  → few urban flux sites/measurements in the US


Sources
Sinks
X
Atmospheric production
Atmospheric loss
Emission
Deposition

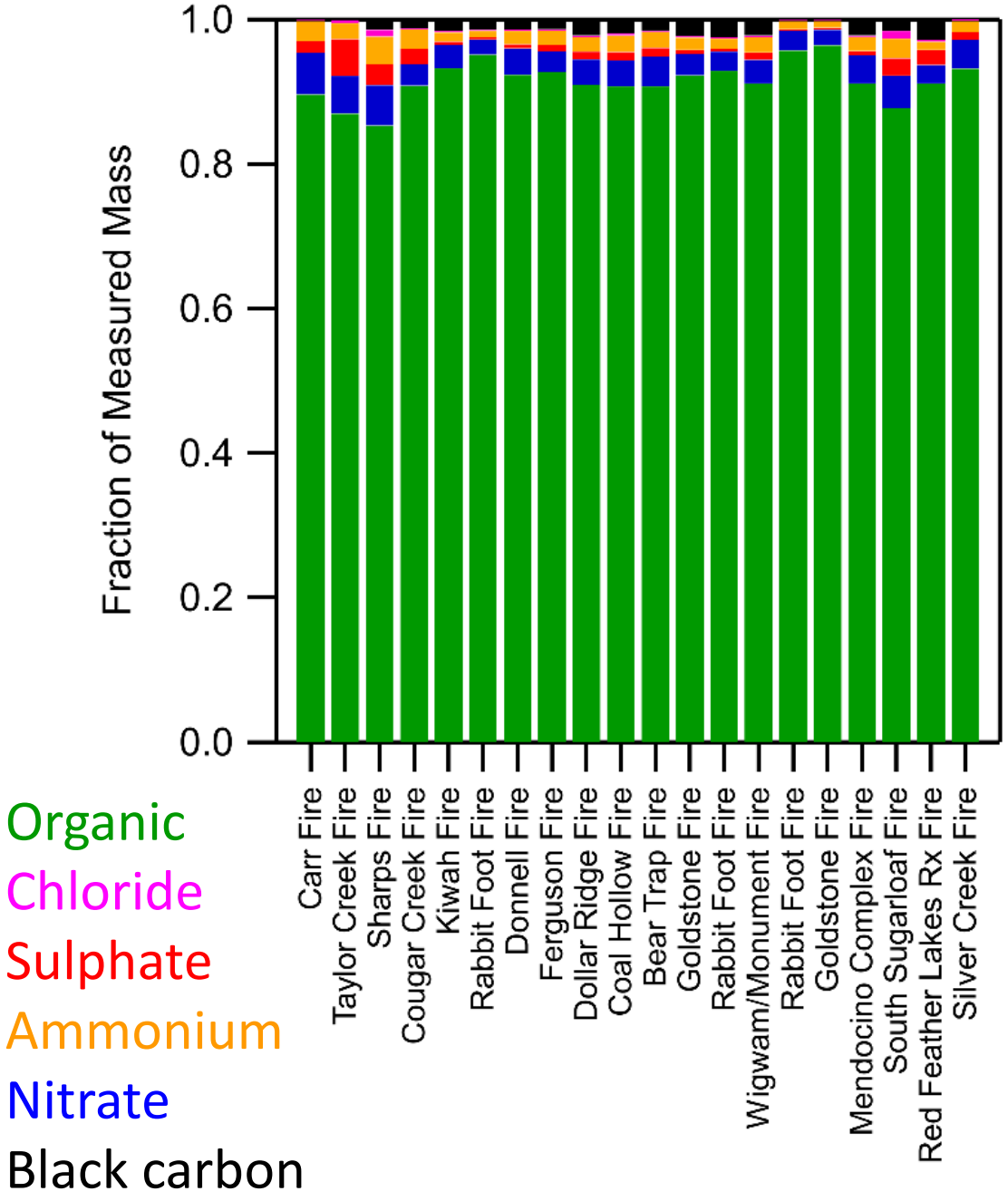
The Western Wildfire Experiment for Cloud Chemistry, Aerosol Absorption & Nitrogen



WE-CAN was led by **Emily Fischer** (CSU)
HR-AMS [coPI **Sonia Kreidenweis**, postdoc **Lauren Garofalo** + PhD student **Matson Pothier**]

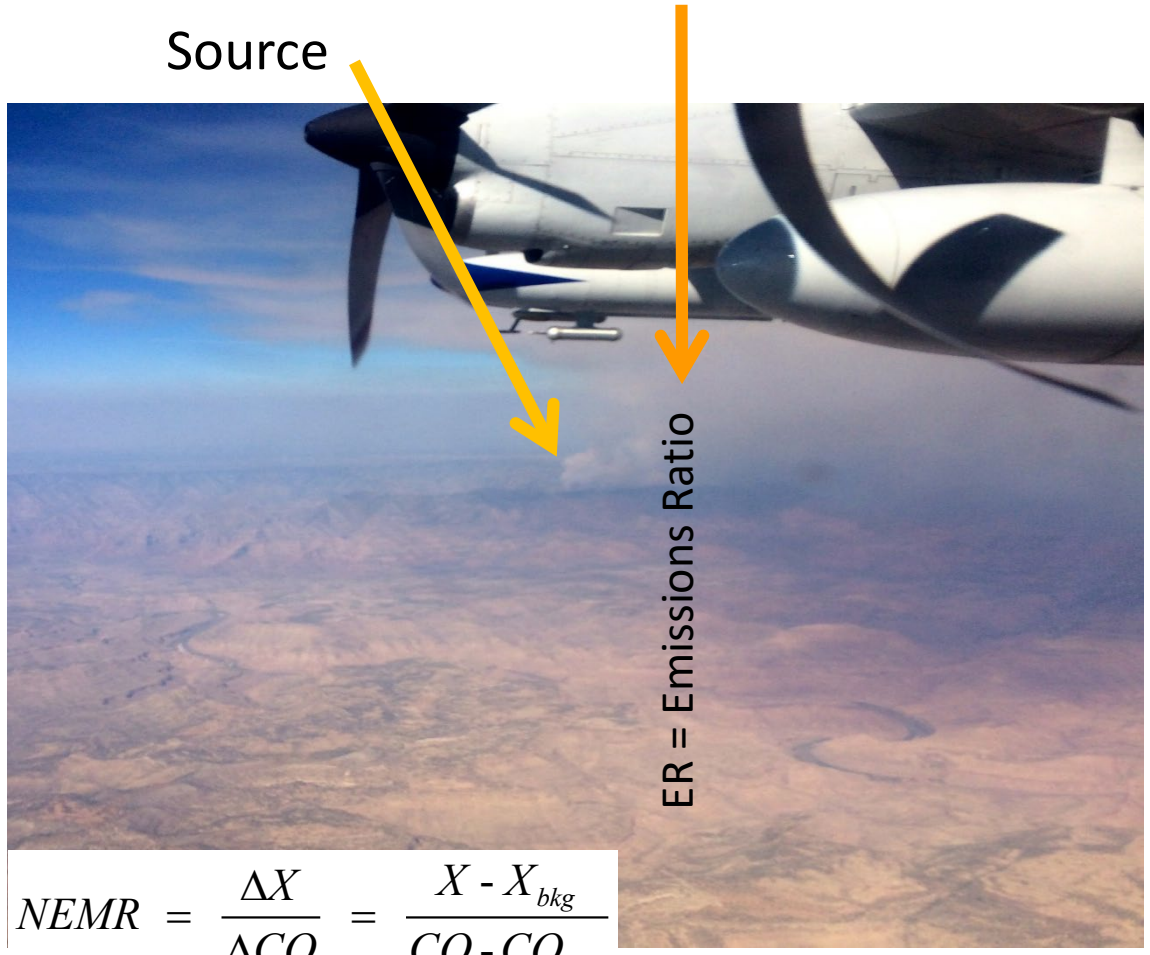
Sub-micron aerosol in wildfire smoke is overwhelmingly organic

AMS provides bulk sub-micron measurements and some useful markers for specific molecules, along with factor analysis for characteristic components

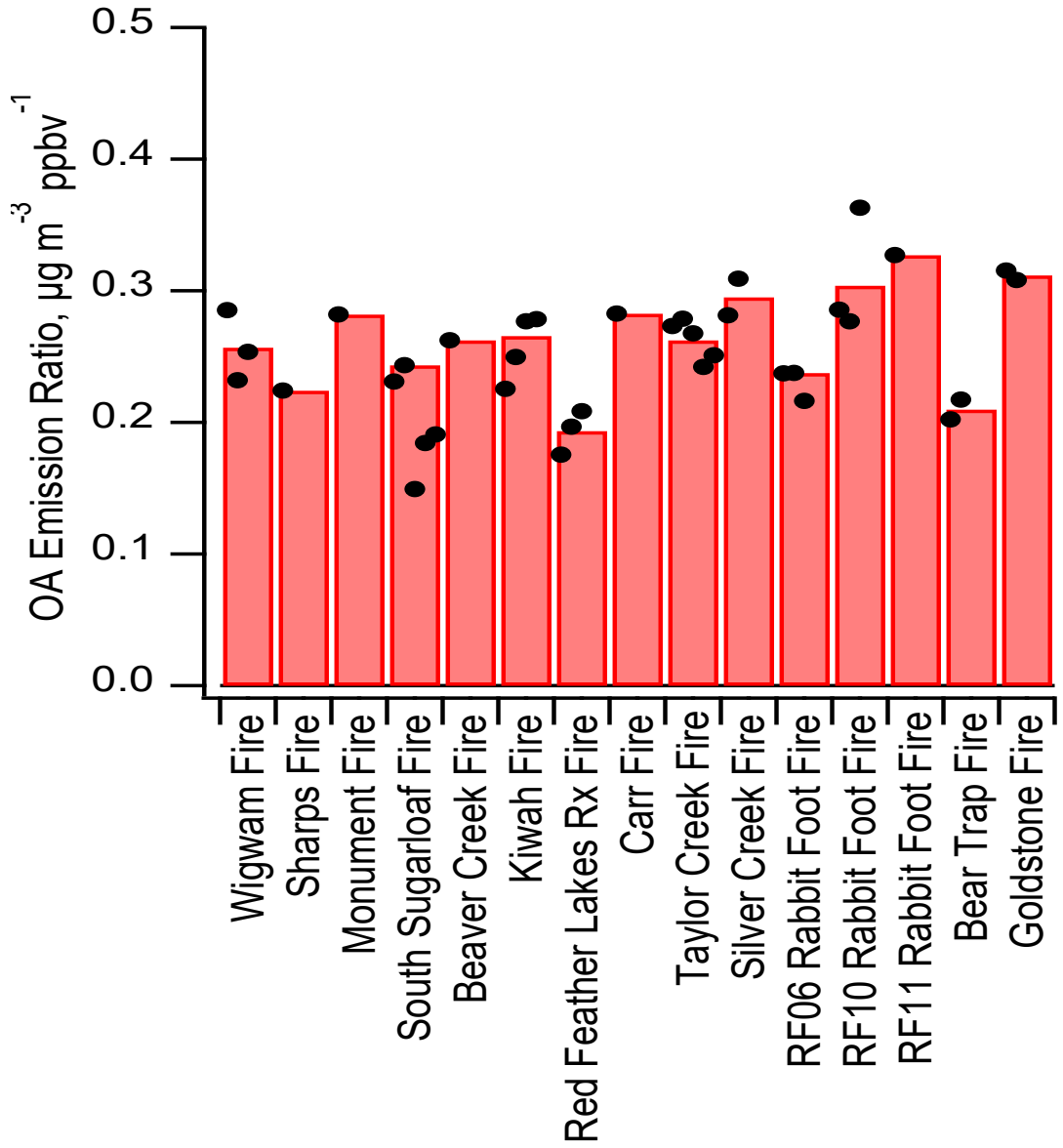


[Garofalo et al. ACS Earth & Space Chemistry. 2019]

We can calculate emissions ratios, accounting for simple dilution, and find that organic aerosol emissions are relatively consistent

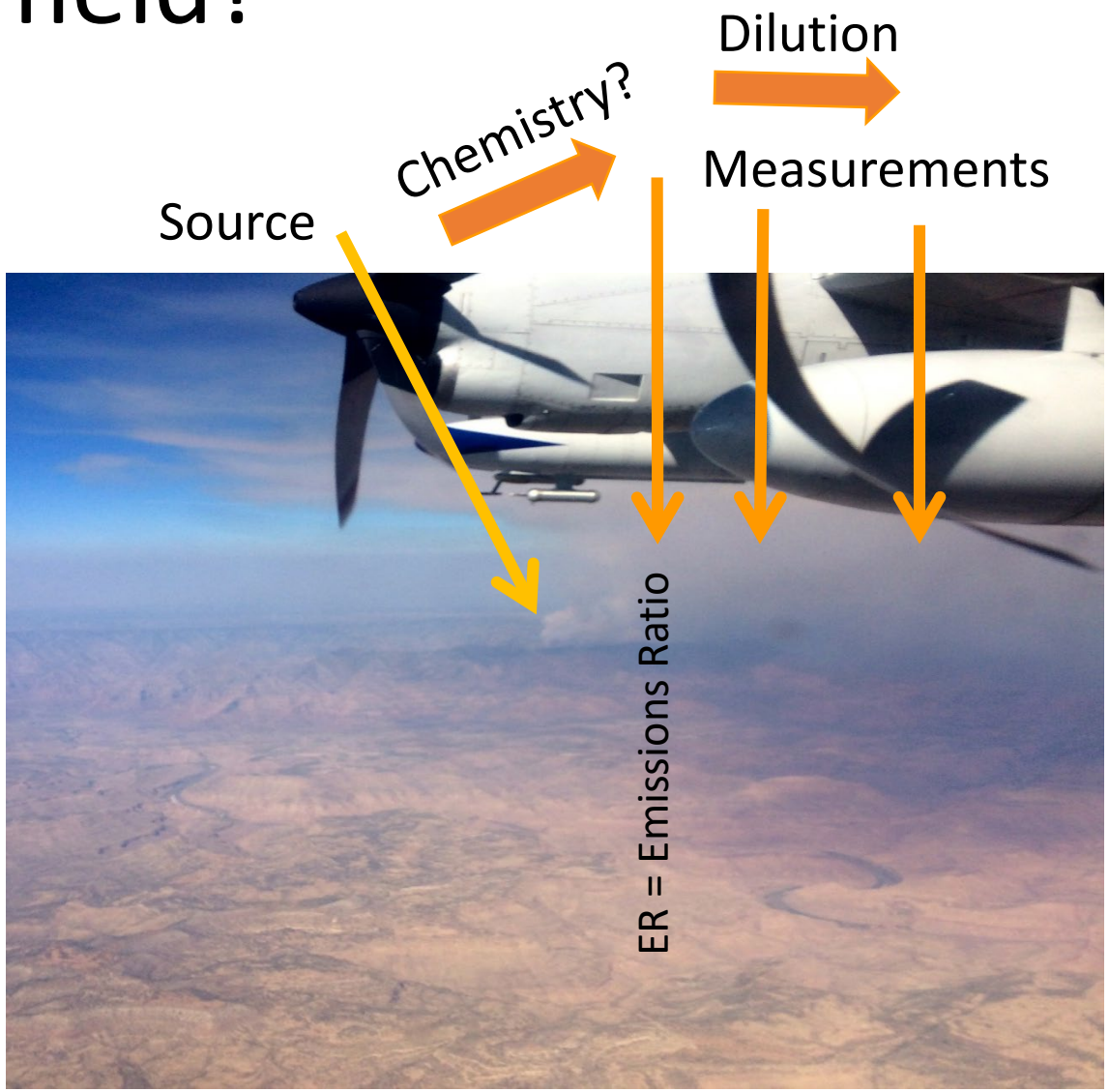


$$NEMR = \frac{\Delta X}{\Delta CO} = \frac{X - X_{bkg}}{CO - CO_{bkg}}$$

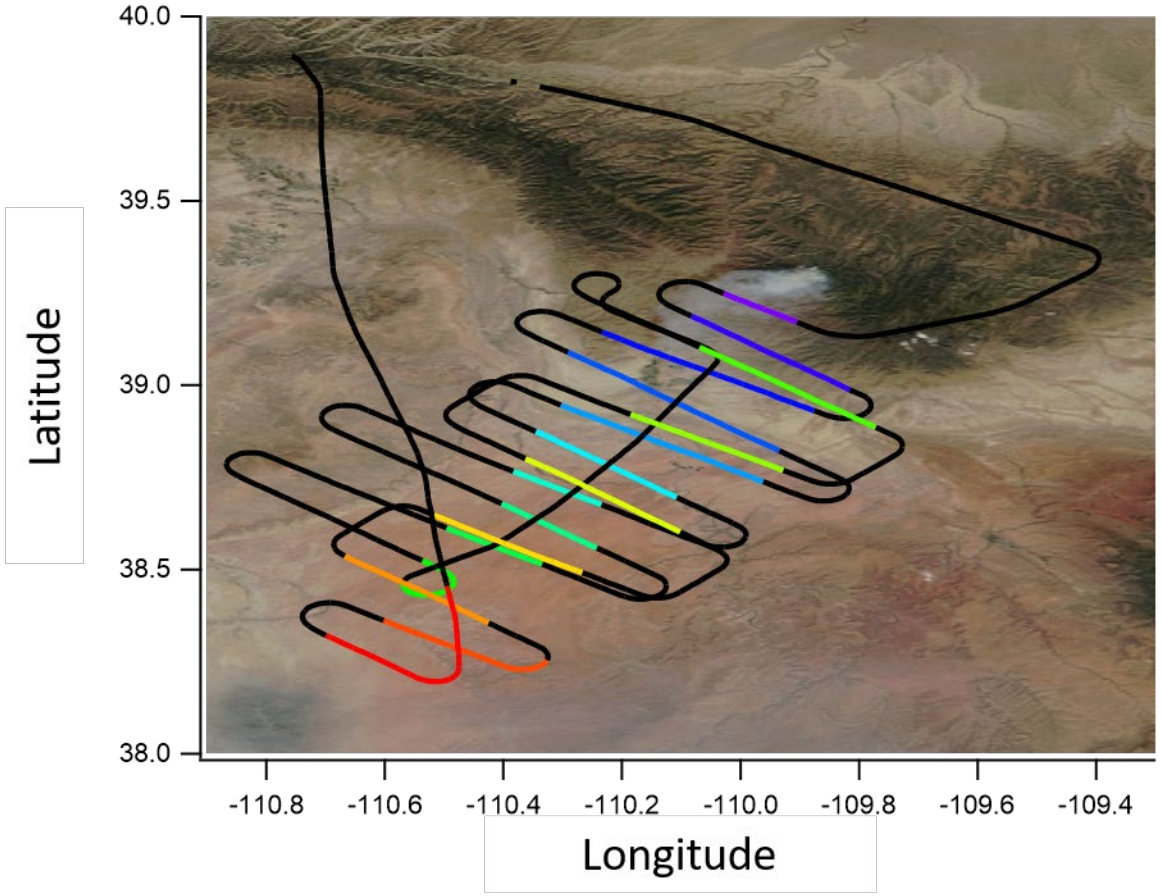


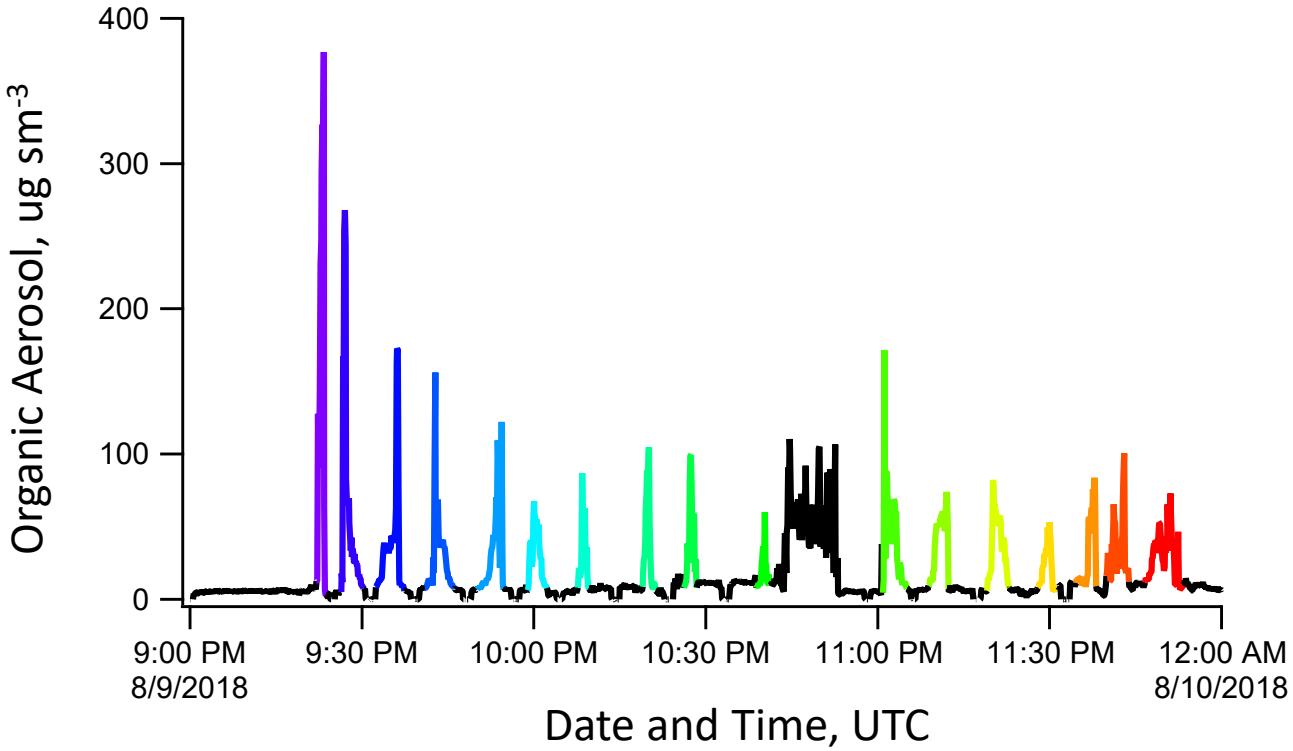
[Garofalo et al. ACS Earth & Space Chemistry. 2019]

Does the fire plume chemically evolve in the near-field?



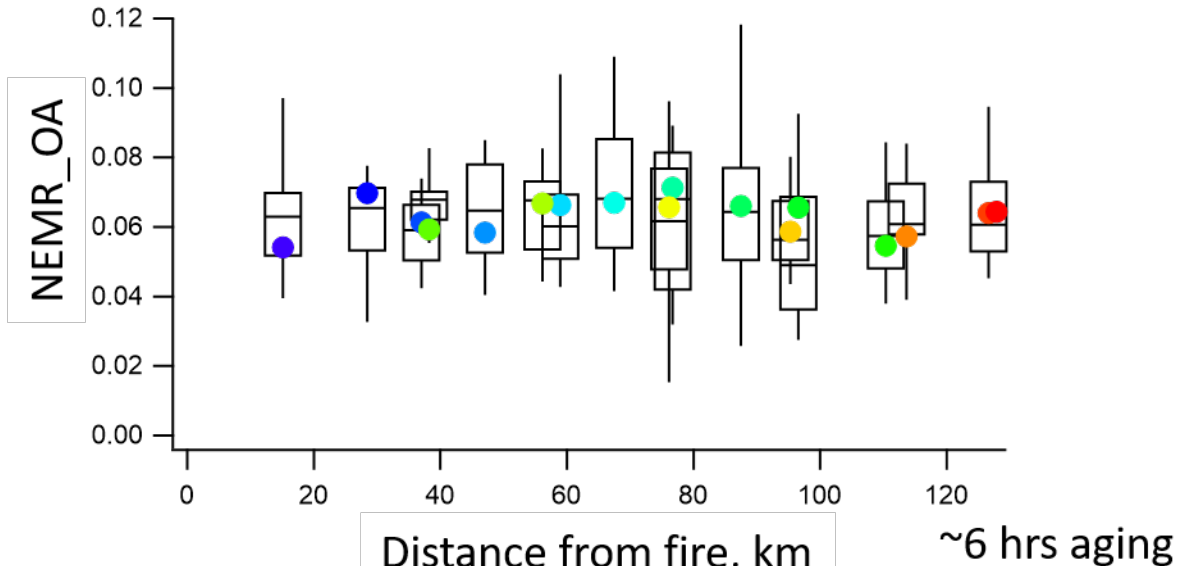
Does the fire plume chemically evolve in the near-field?





RF09: Bear Trap Fire, Utah
Pine + Aspen

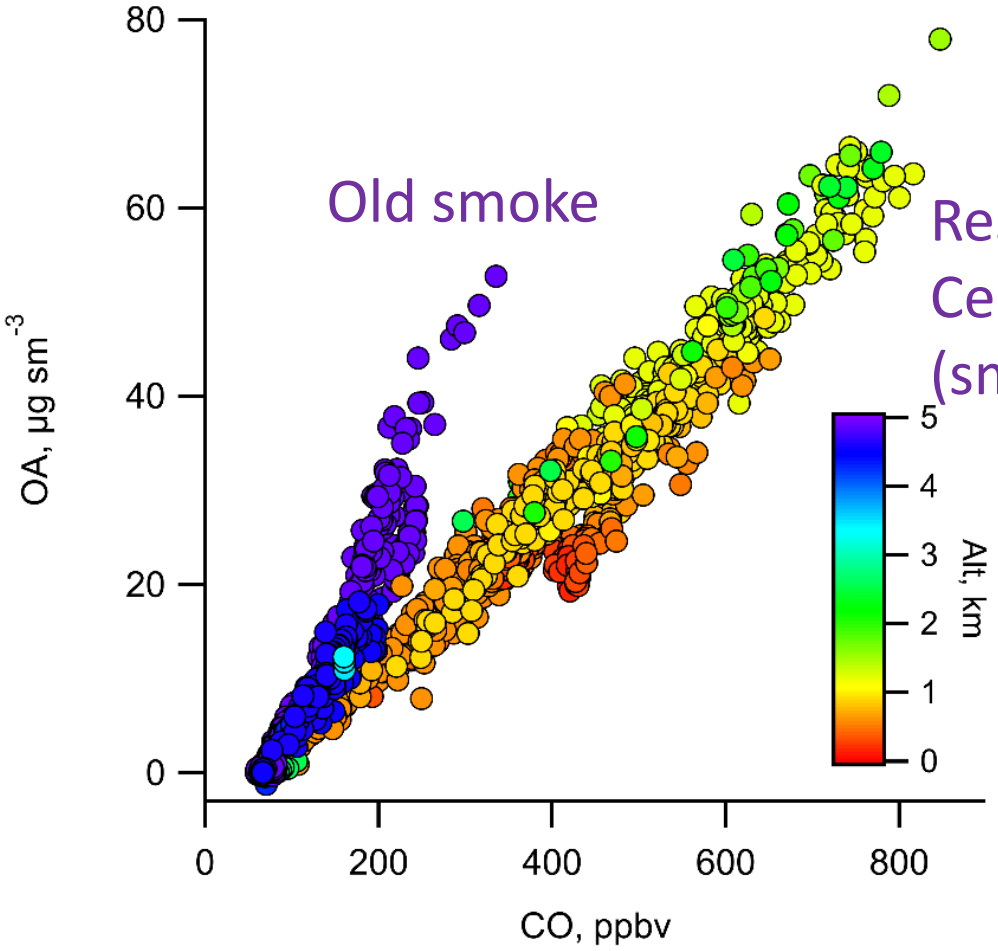
OA changes: oxidation & dilution-driven evaporation balance!

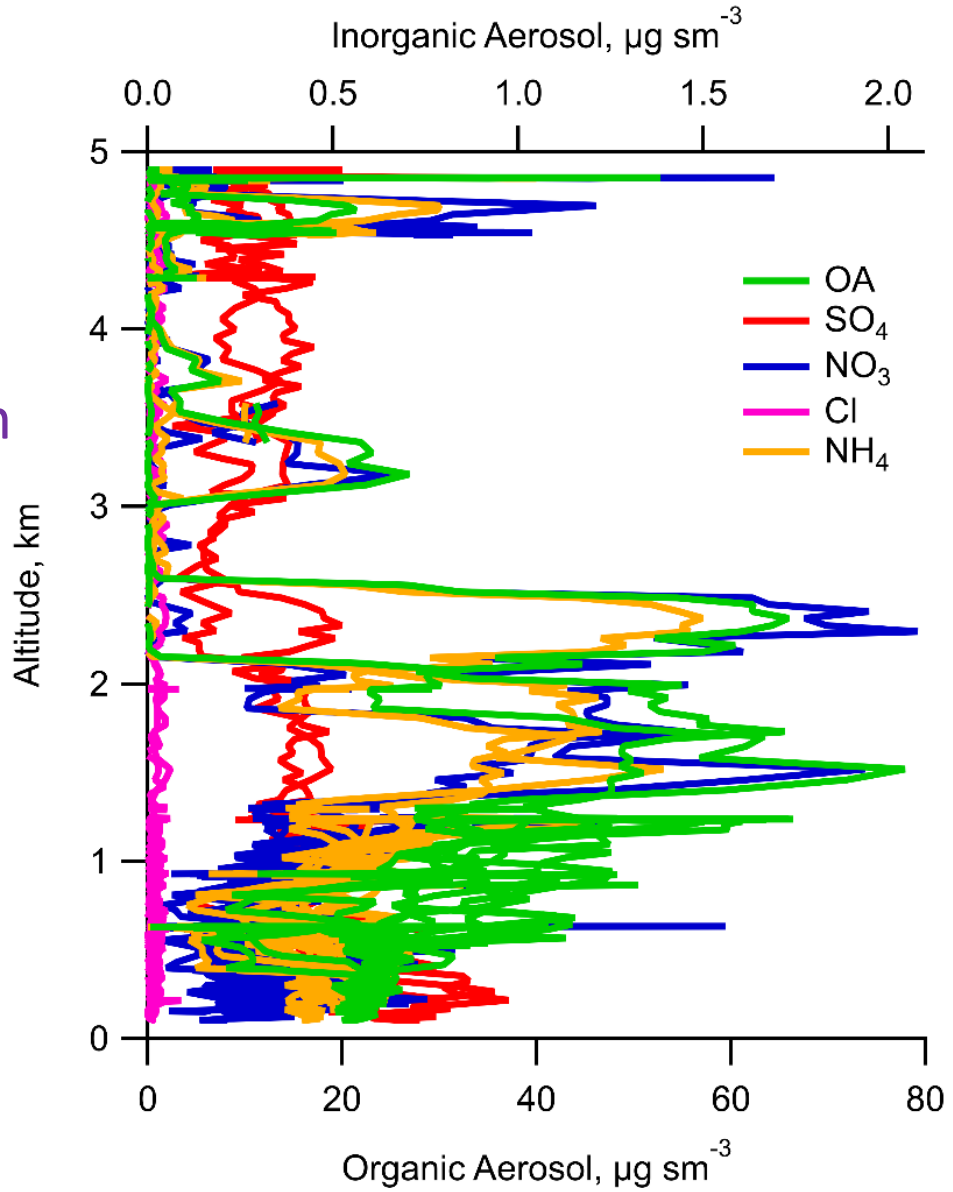


~6 hrs aging

[Garofalo et al. ACS Earth & Space Chemistry. 2019]

What happens on longer timescales and in polluted environments?

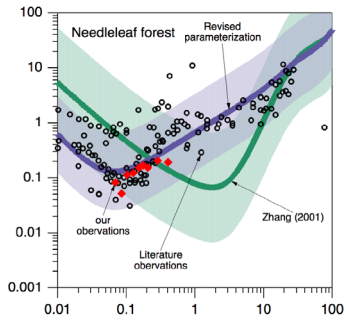




What else would have been useful for this type of work?

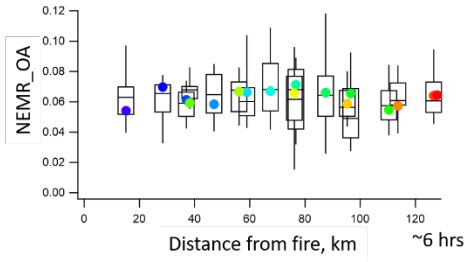
- Size distribution measurements are not trivial
- Organic aerosol molecular speciation can compliment bulk aerosol composition
- Refractory inorganic ions provide additional information
- Understanding of volatility (PMF can only get you so far, although thermal denuders have time resolution challenges)
- AMS only measures the non-refractory component of $PM_{0.8\text{-}800}$ – what about the rest of the aerosol?

Observational constraints on emission and deposition terms are useful – and provide insight on chemistry



Dry deposition is an important – but poorly characterized – loss process for sub-micron particles

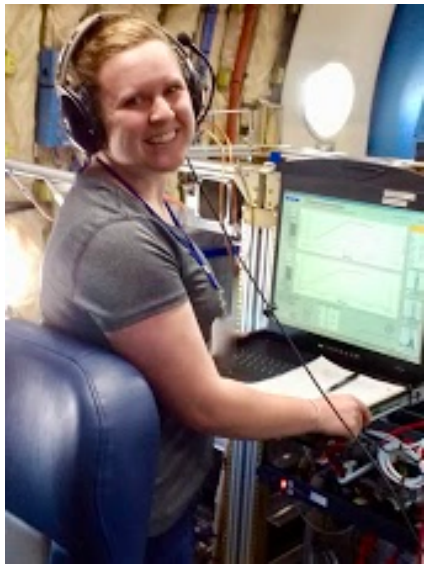
Flux measurements provide insight on both sources and sinks of trace gases and particles



Organic aerosol is complex: chemical tracers are useful for separating out processes

**But it's important to recognize measurement limitations*

Acknowledgements

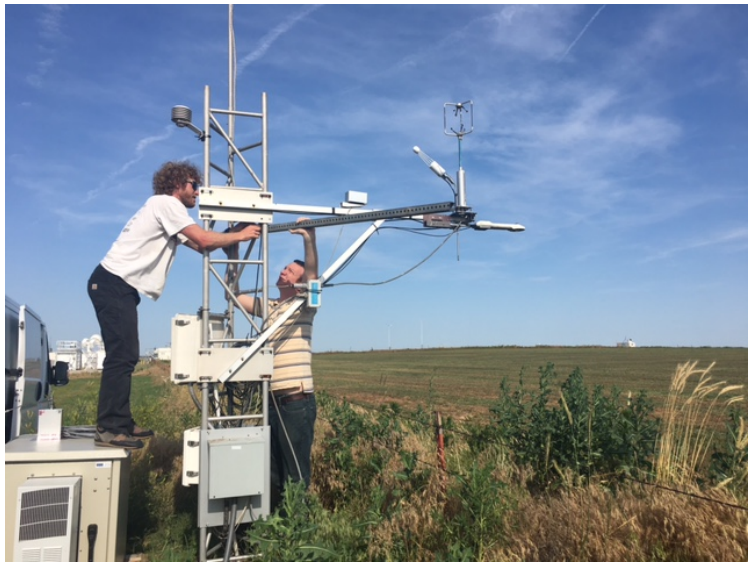


Dr. Lauren Garofalo

Matson Pothier

+ Prof. Sonia Kreidenweis (coPI), Dr. Ezra Levin (CSU, black carbon), Dr. Teresa Campos (NCAR for CO measurements) and the entire WE-CAN Science Team

+ Holly DeBolt (CSU, aerosol fluxes)



Ethan Emerson & Gavin McMeeking [Handix]