Laboratory Water Uptake Studies of Biomass Smoke and Assessment of BC Parameterization in DOE Climate Model

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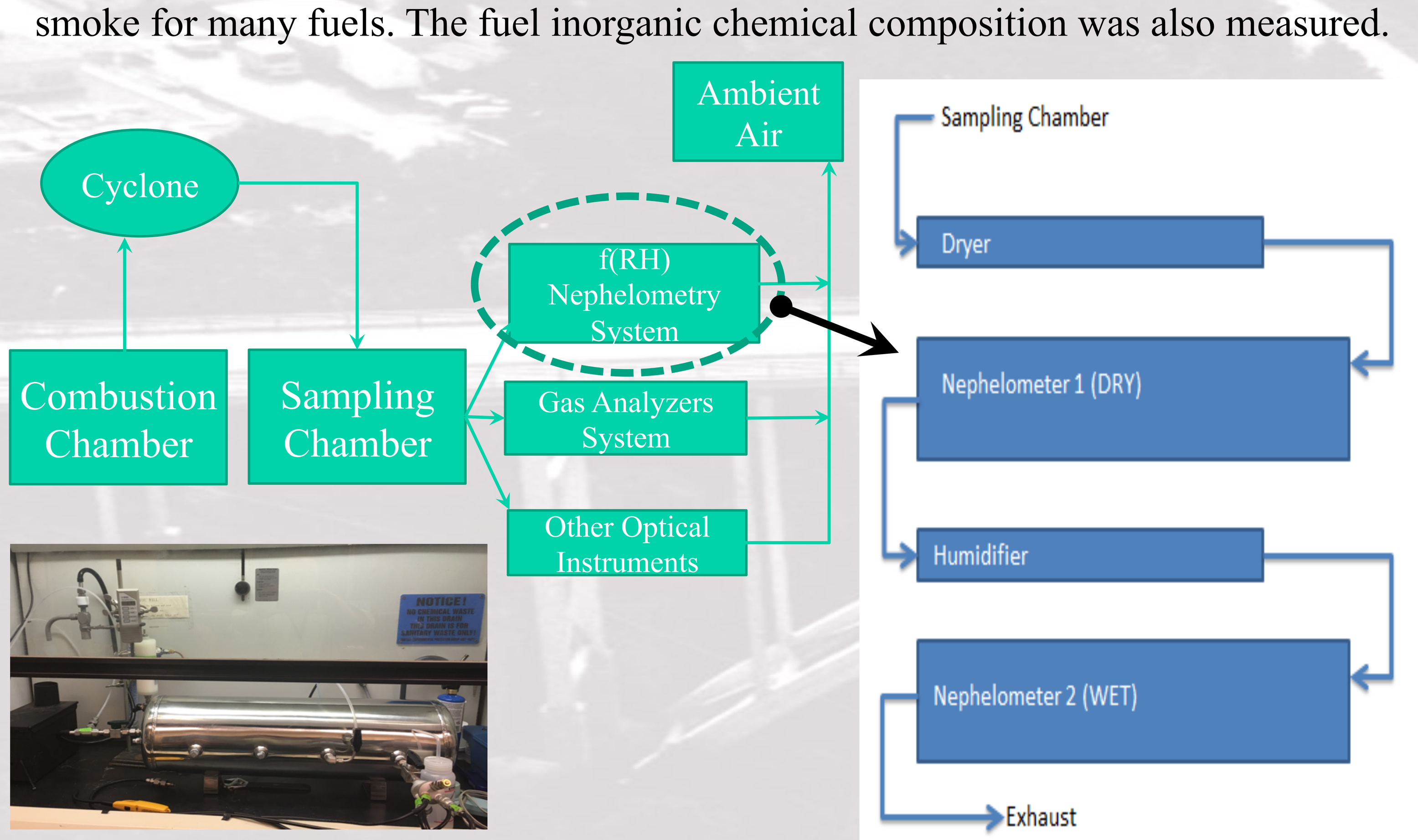
1. Abstract

Aerosol interactions with water control the evolution of optical properties and their cloud condensation nuclei activity that affect the Earth's radiative budget. Water uptake increases light scattering by aerosol growth, absorption by black carbon by lensing, cloud reflectivity, and lifetime and aerosol scavenging with competing effects on climate forcing. Model parameterizations must capture process details including composition of fresh emissions and the evolution of mixing state as they age. We report laboratory studies of fresh biomass burning smoke from Southwest US fuels. We measured a suite of aerosol properties with a focus on aerosol light extinction and their hygroscopic response. While optical properties were strongly dictated by the flaming versus smoldering nature of the burn, we find that aerosol hygroscopicity is intimately linked to fuel type and its ion content. We also analyzed the treatment of aerosols by the global CAMChem model. In particular, sensitivity of BC in MAM4 where it is transferred from the primary Aitken mode to the accumulation mode after accumulating 8 monolayers of material on aging is evaluated by varying this threshold from from 1 to 12 monolayers. We compare the timescales of this BC conversion in CAMChem that range from hours to several days to those in a more detailed BC mixing-state model PartMC MOZAIC. We examine the effects of parameters on global distributions of BC to assess their importance and develop a strategy to constrain them with field data.

2. Experimental Methods

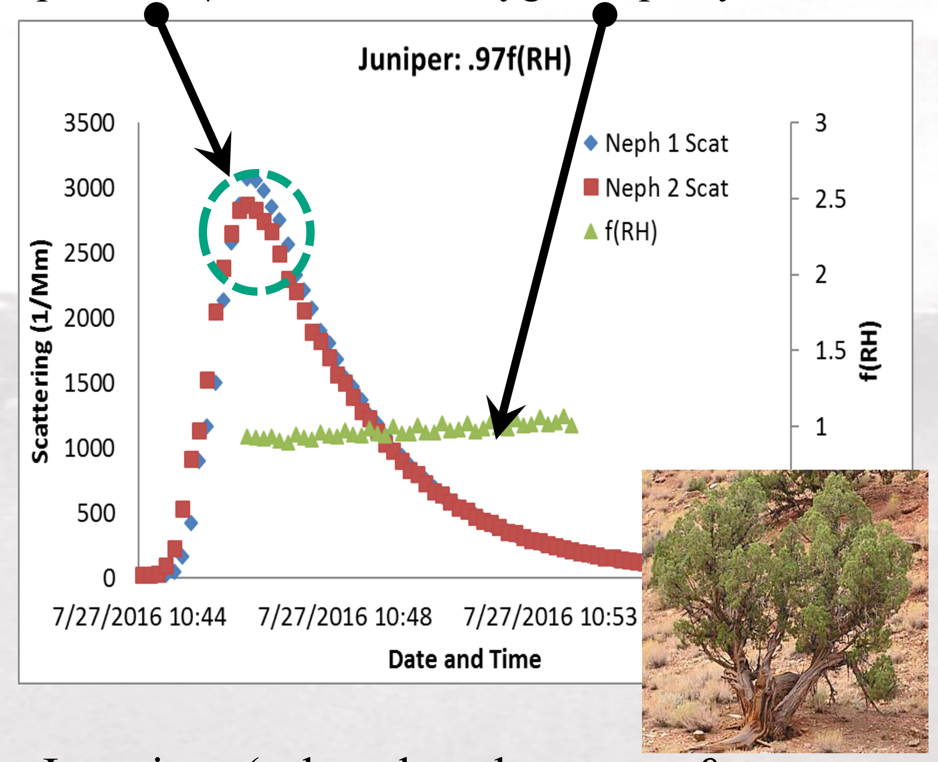
Light scattering by particles was measured using a **controlled RH nephelometry system** (Ecotech Aurora, 450 nm). Smoke emissions were sampled under a fume hood through a cyclone into 9-gallon stainless mixing chamber with minimal dilution. Scattering at 85% RH and for dry conditions was measured to deriver f(RH) for smoke for many fuels. The fuel inorganic chemical composition was also measured.



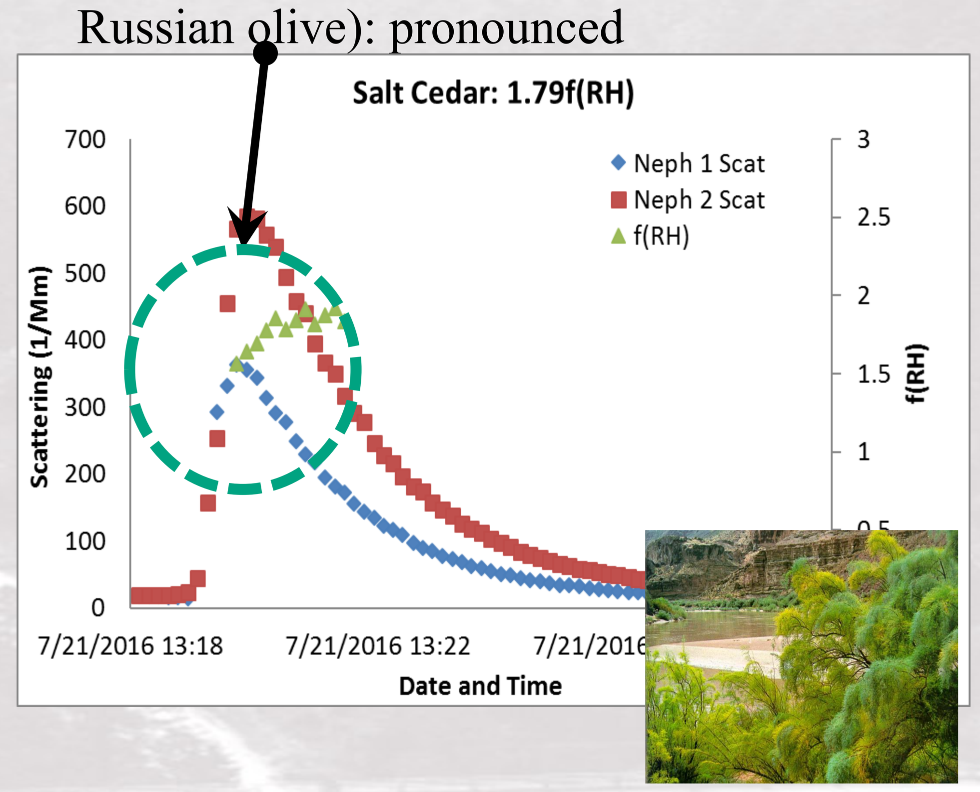
3. Results: Scattering f(RH)

Evidence of particle collapse with ↑RH

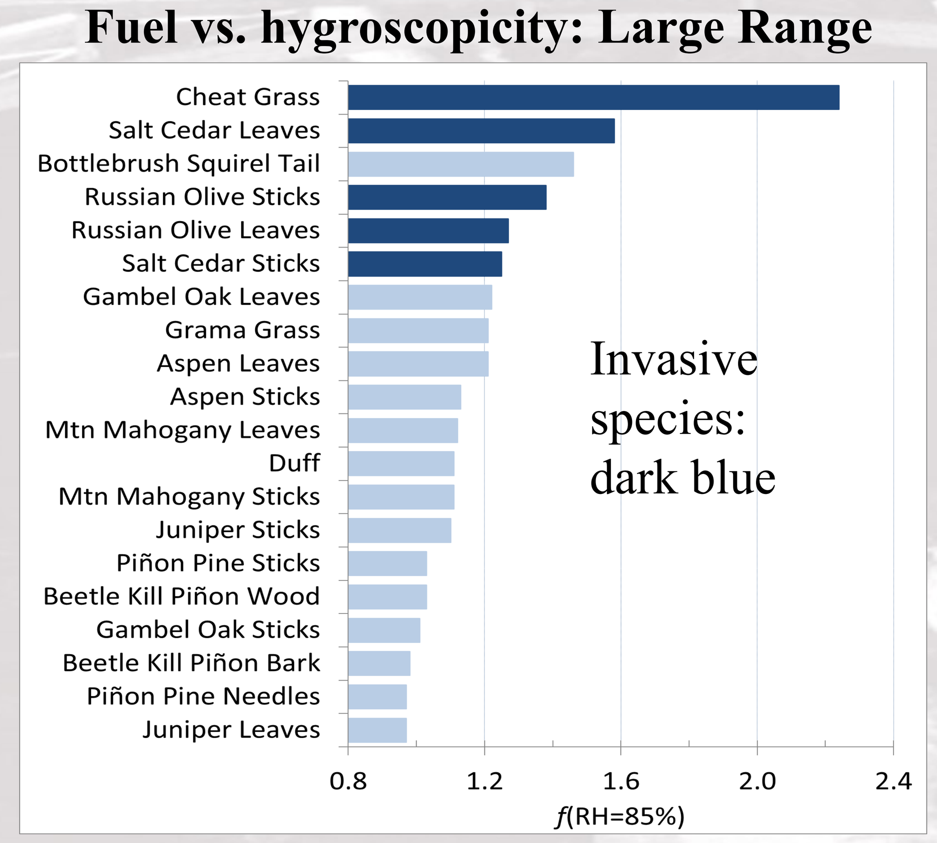
Pines, juniper, evergreens show low hygroscopicity.



Invasives (salt cedar, cheatgrass & Russian olive): pronounced



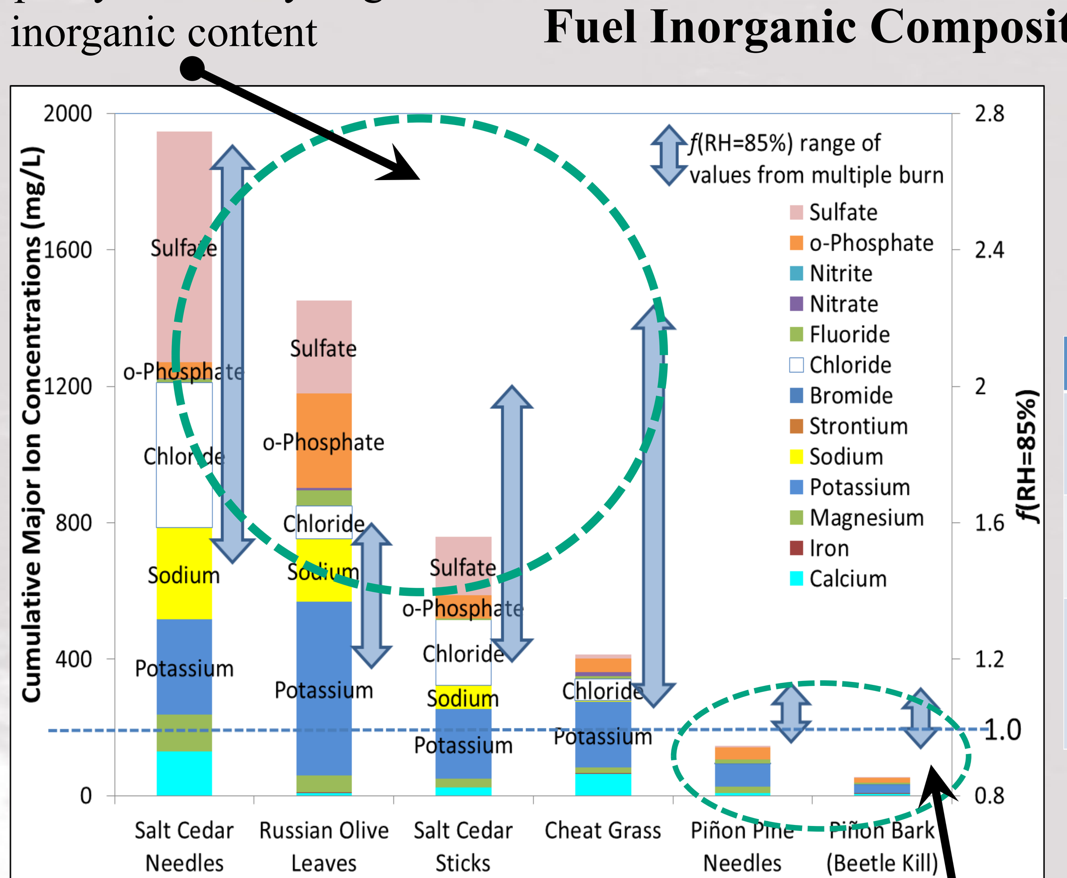
Fuel vs. hygroscopicity: Large Range



4. Results: Fuel Chemistry

Hygroscopicity driven by large fuel inorganic content

Fuel Inorganic Composition



Weakly Hygroscopic: OC dominated

5. Predictive Framework

Fuel Type	Smoldering	Flaming
Evergreen Needles or Woods	Nearly Hydrophobic	Slightly Hygroscopic
Deciduous Woods	Weakly Hygroscopic	Moderately Hygroscopic
Grasses, Leaves or Needles with Large Inorganic Fraction	Strongly Hygroscopic	Extremely Hygroscopic

Increasing hygroscopicity

Hygroscopicity is driven by:

1) Fuel inorganic content is key
2) Rules on smoke hygroscopicity:
   - More Inorganic > Less Inorganic
   - Flaming > Smoldering
   - Leaves > Woods and Barks
   - Deciduous > Evergreen
   - Invasives > Natives

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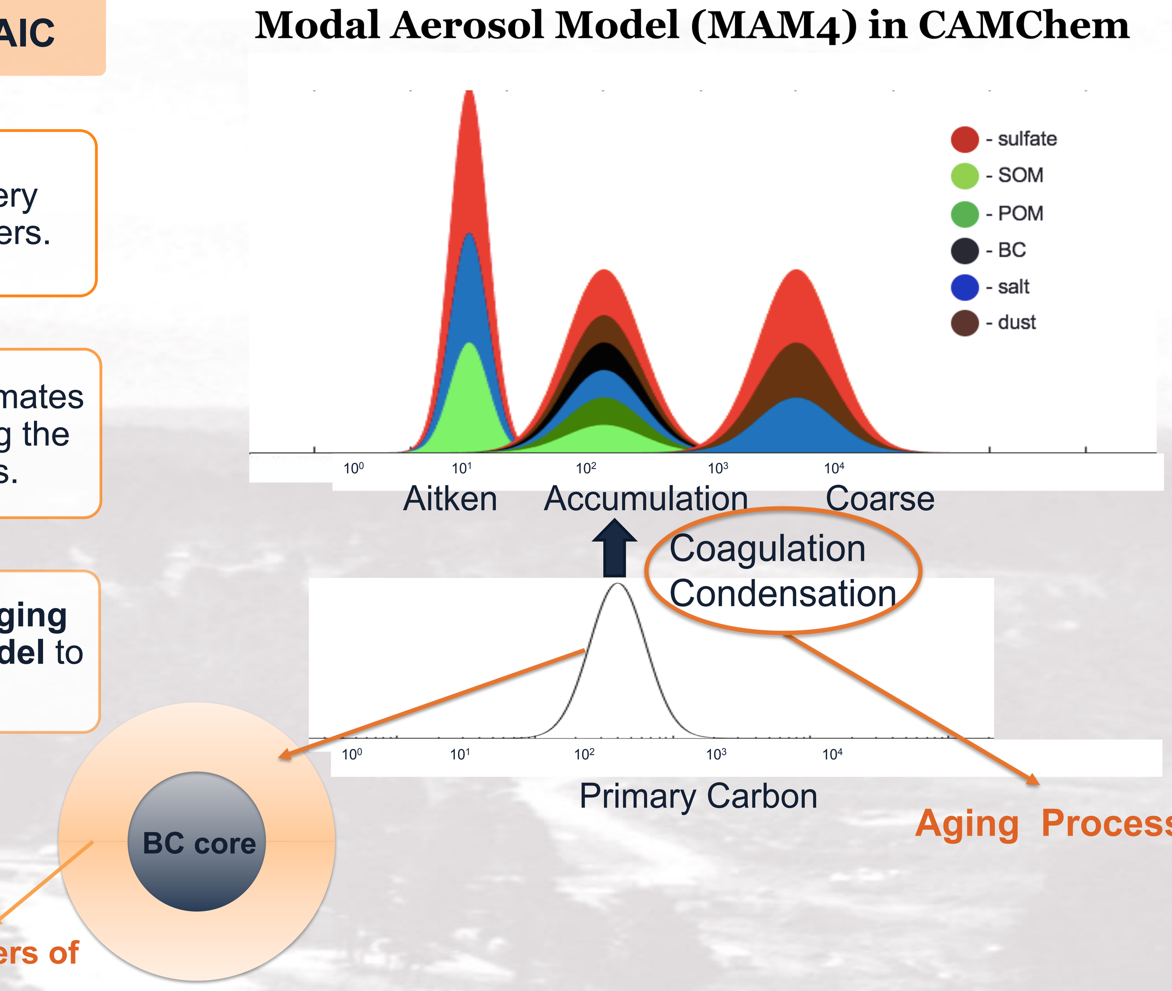
6. Assess BC Aging Mechanism in CAMChem

Goal: Assess Aging in CAMChem & PartMC MOSAIC

- CAMChem uses **mechanistic** aging rates, very **sensitive** to the choices of assumed parameters.
- Particle resolved model (PartMC-MOSAIC) estimates BC aging timescales **more precisely** by tracing the mass and composition of individual particles.
- PartMC-MOSIAC **parameterization of BC's aging** can be applied to the **output of CAMChem model** to assess the accuracy of its aging criterion.

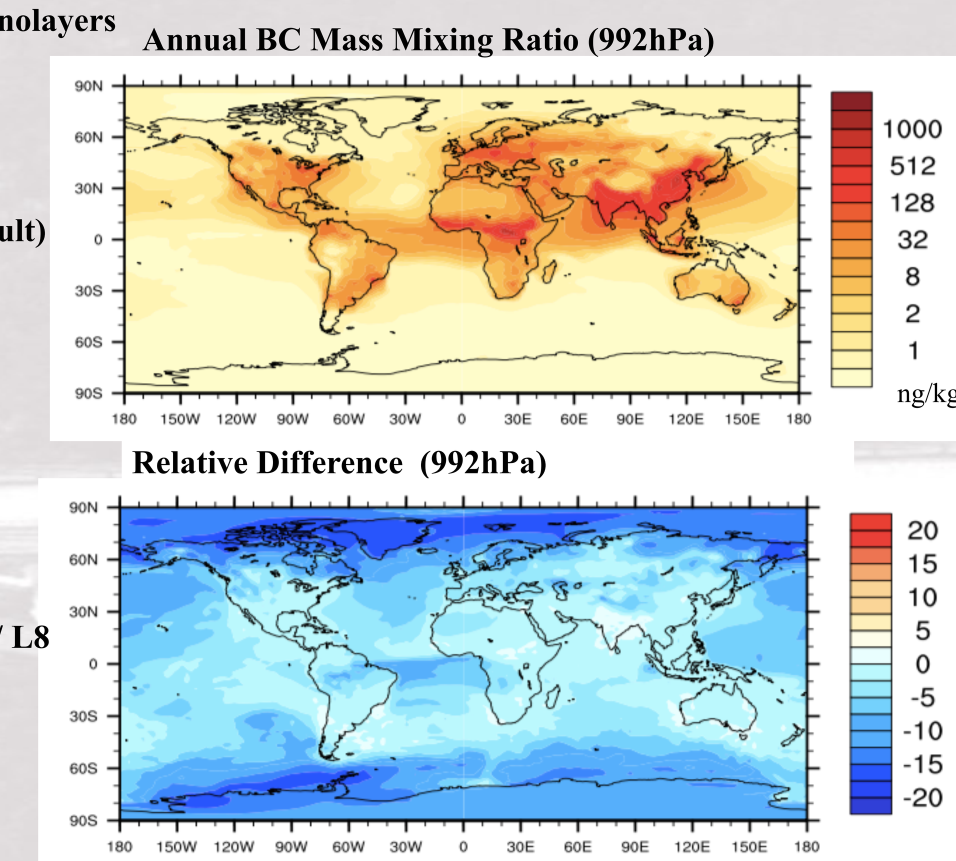
The equivalent of 8-monolayers of sulfate is needed to transfer material from the primary carbon mode (fresh) into the accumulation mode (aged).



7. Sensitivity to Monolayer Criterion

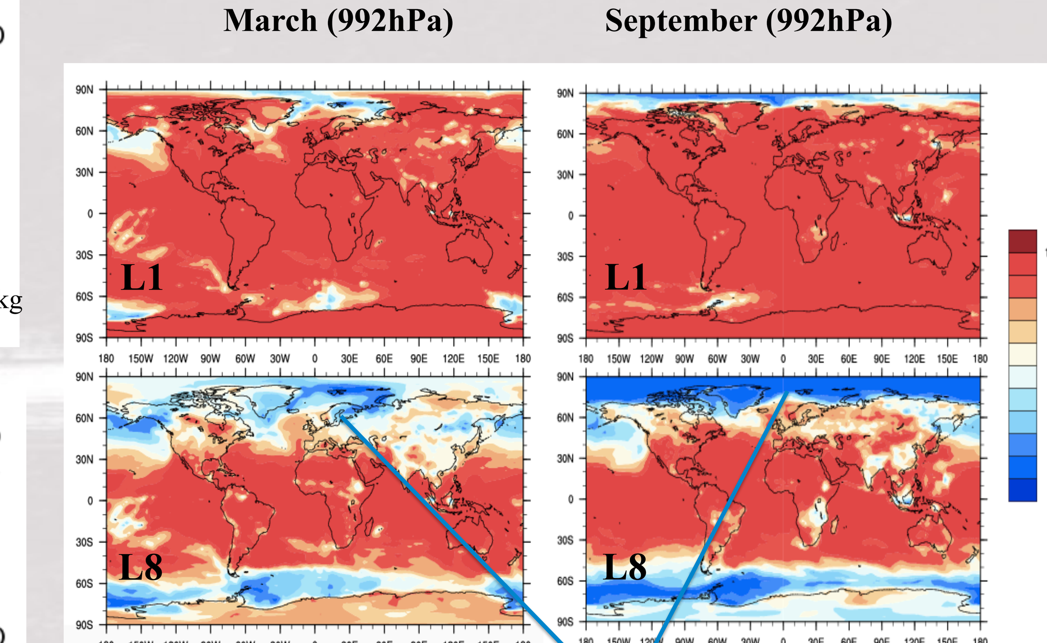
BC Burden

L1: 1 monolayer; L8: 8 monolayers



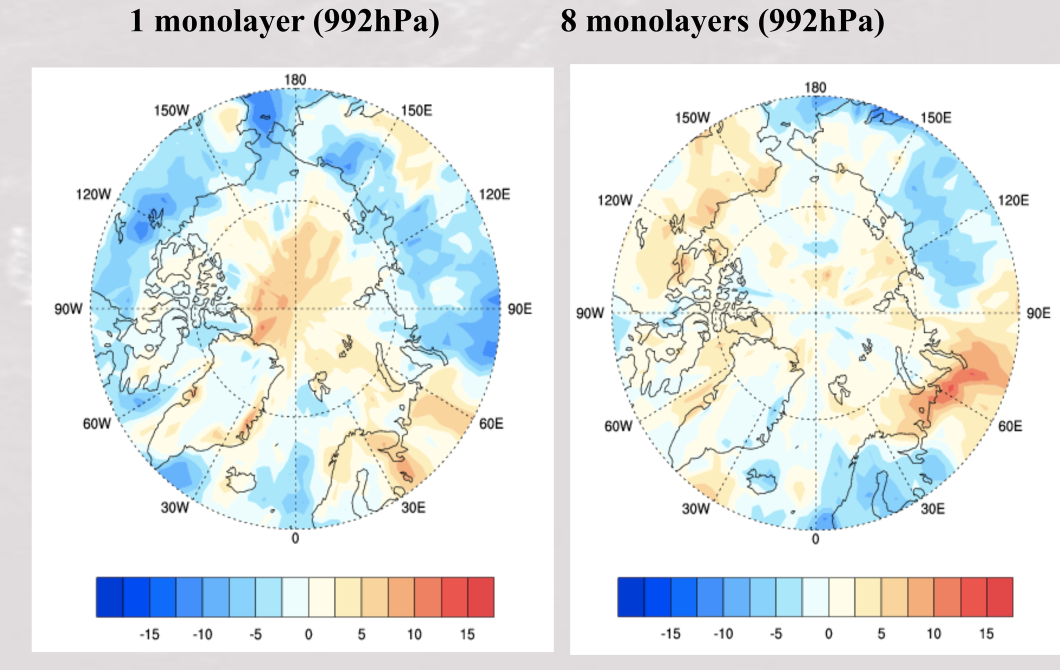
Relative differences highest at high latitudes (distant from sources).

BC Mixing State

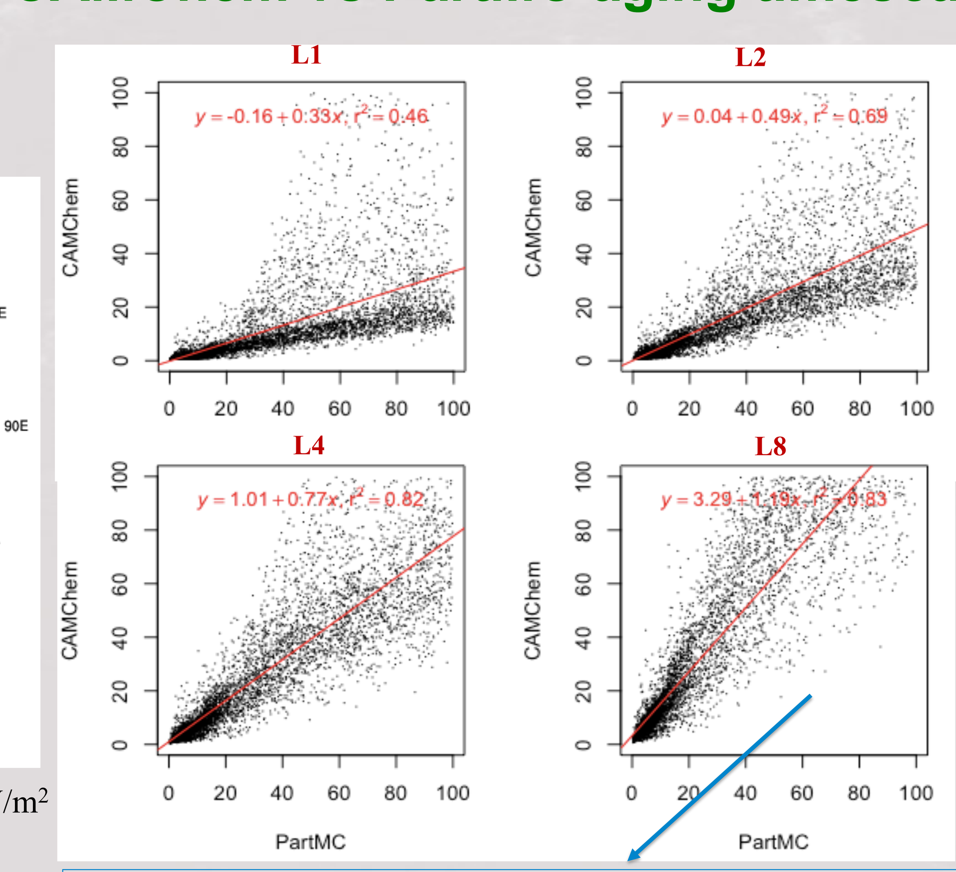


Arctic: Most BC is in primary carbon mode (externally mixed)!

Arctic Annual BC Direct Forcing



CAMChem vs PartMC aging timescale



When using an 8-monolayer sulfate criterion, the aging timescales have the closest linear-regression slope to 1 and the highest R^2 value among four sensitivity cases.

8. Results: BC Models

- BC burden is most sensitive to aging criterion in the high-latitude regions.
- Aging timescales range from less than one hour to several days.
- Condensation of SOA and sulfate plays a dominating role in BC aging, compared to coagulation.
- MAM4 aging timescales are broadly consistent with PartMC-MOSAIC aging timescales.