Uncovering the Air Quality and Climate Impacts of Wildfire Aerosols from Western U.S. Fires

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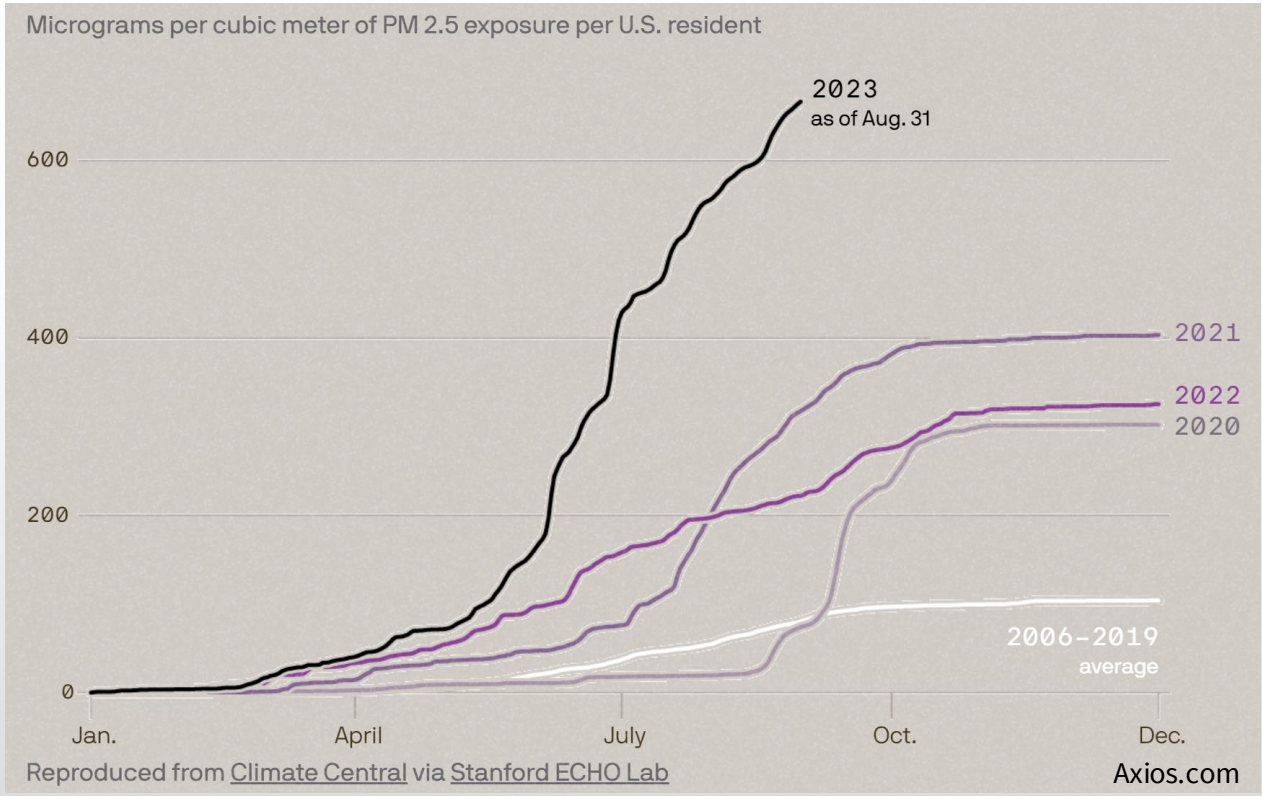
Increased Exposure to Wildfire Smoke

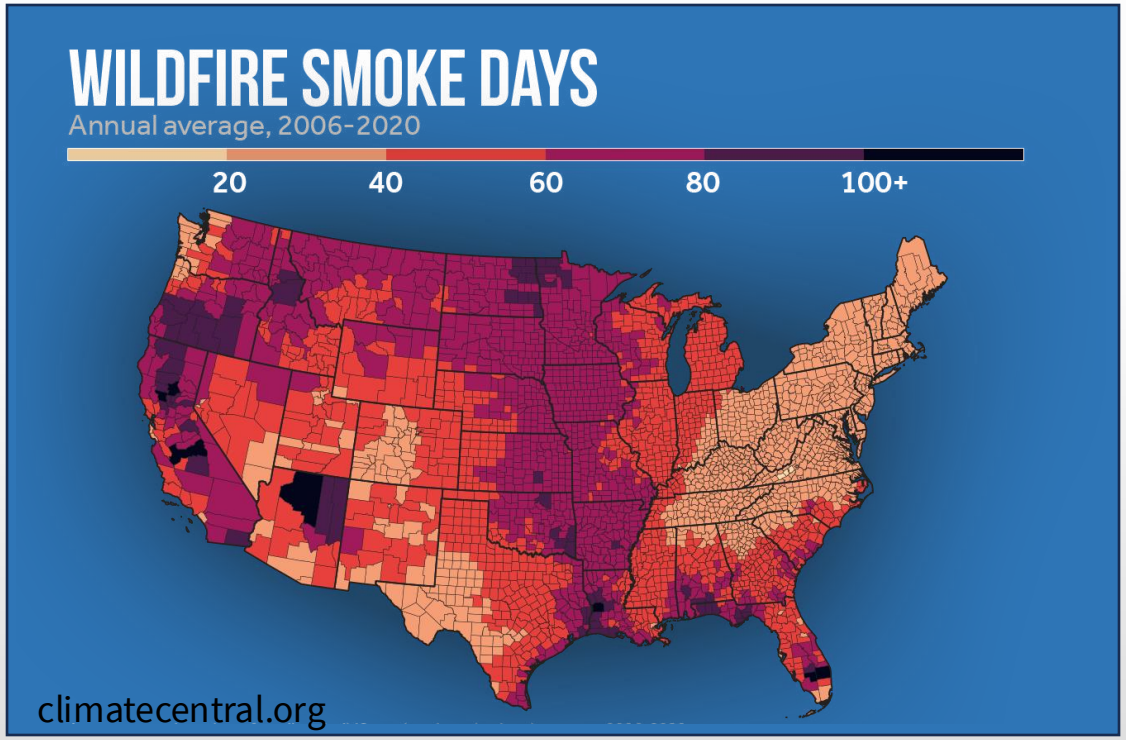
- Increased wildfire events impact both the climate and human health.
- **Short term effects**: Eye, nose, throat, and lung irritation, runny nose, coughing and sneezing.
- **Long term effects**: Asthma, chronic bronchitis, cardiovascular damage, and increased mortality in susceptible populations.



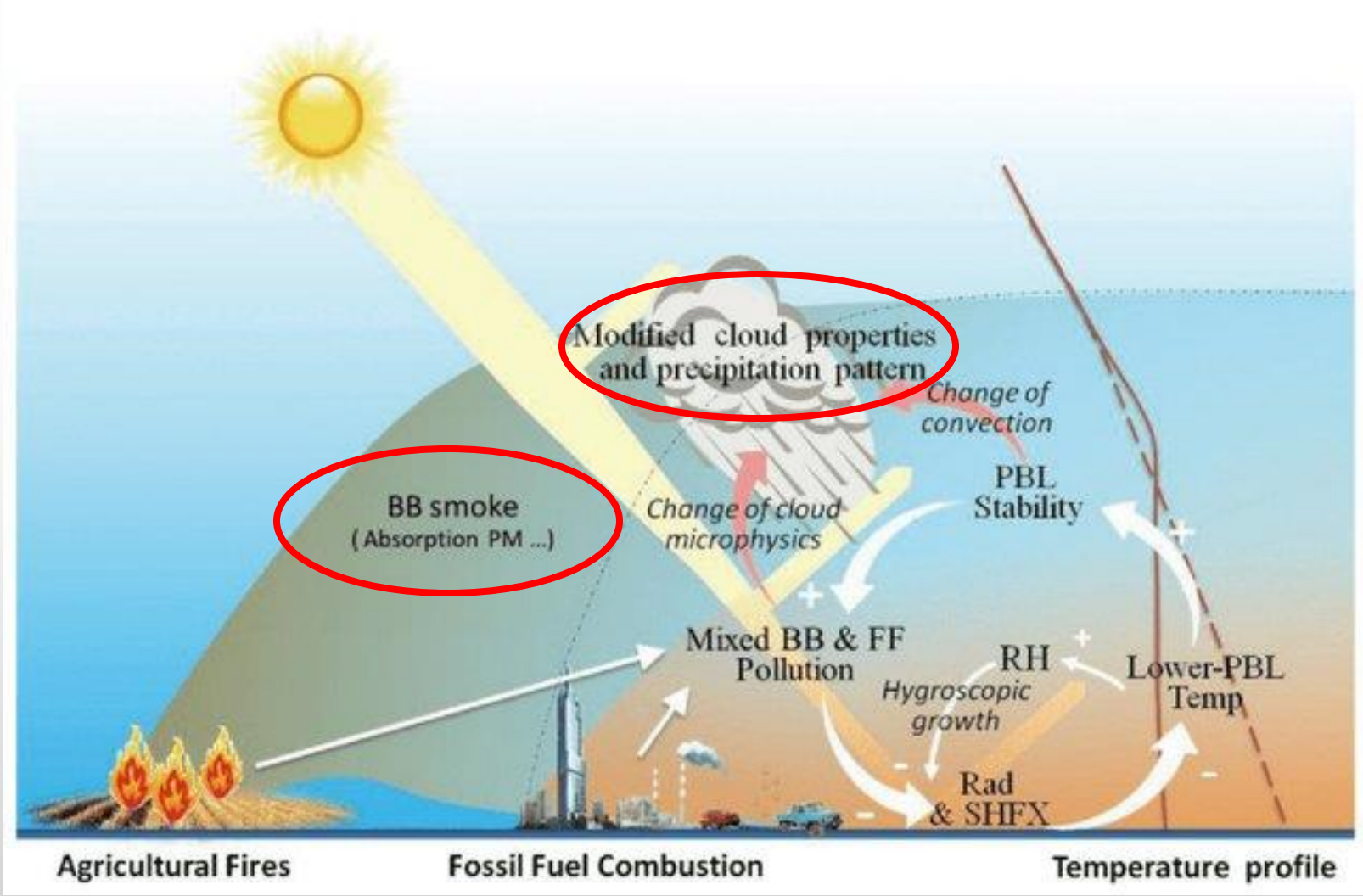
Cumulative smoke-related pollution exposure

Average Wildfire Smoke Days

- Smoke days—defined as a day with overhead smoke measured through satellites.
- Wildfire in the contiguous U.S. mostly comes from the western part and Canada.
- Dark spot in Kansas is Flint Hills—has a lot of prescribed burns.

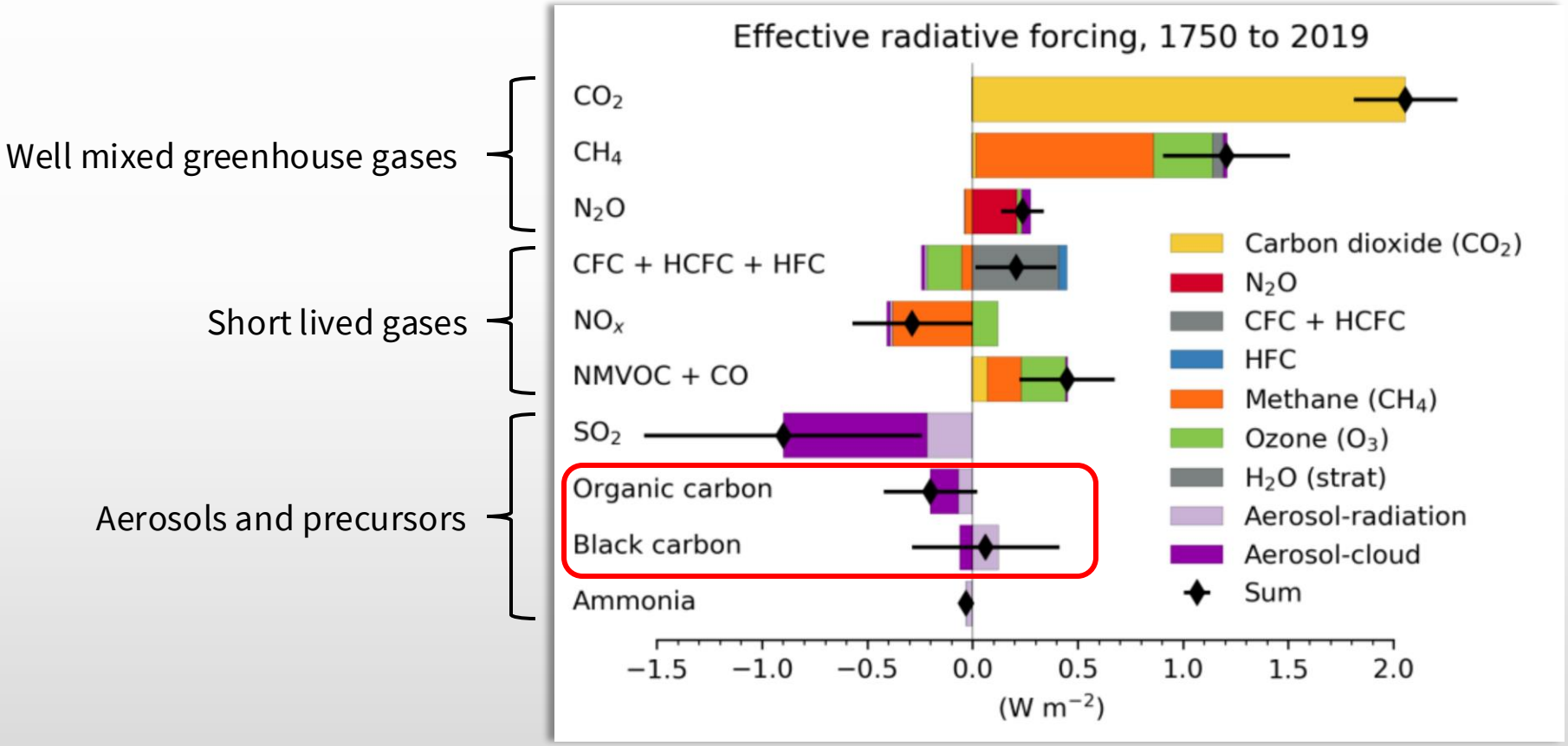


Smoke Impacts on Climate



Meng et al., 2019, Sci. Chi. Earth Sci.

Radiative Impact of Pollutants



Naik et al., 2021, IPCC

<u>Black Carbon (BC)</u>:

- An optical definition for soot
- Large errors in radiative forcing

<u>Organic Carbon (OC)</u>:

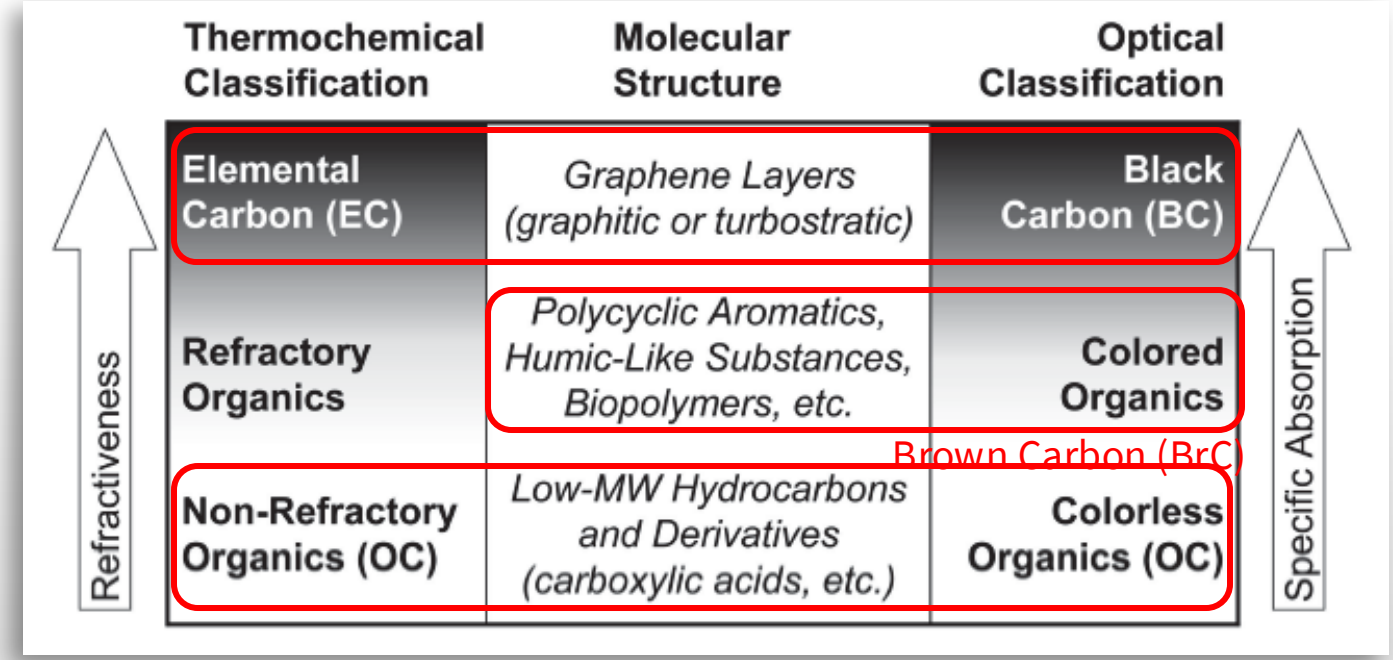
- Low volatility organic material
- Low/No absorption at long visible wavelengths

Carbonaceous Aerosol Classifications

- Carbonaceous aerosols are generated during incomplete combustion: biomass and fossil fuel burning

- Black carbon (BC):
  - Predominantly emitted during flaming
  - Refractory material
  - Strong visible light absorption

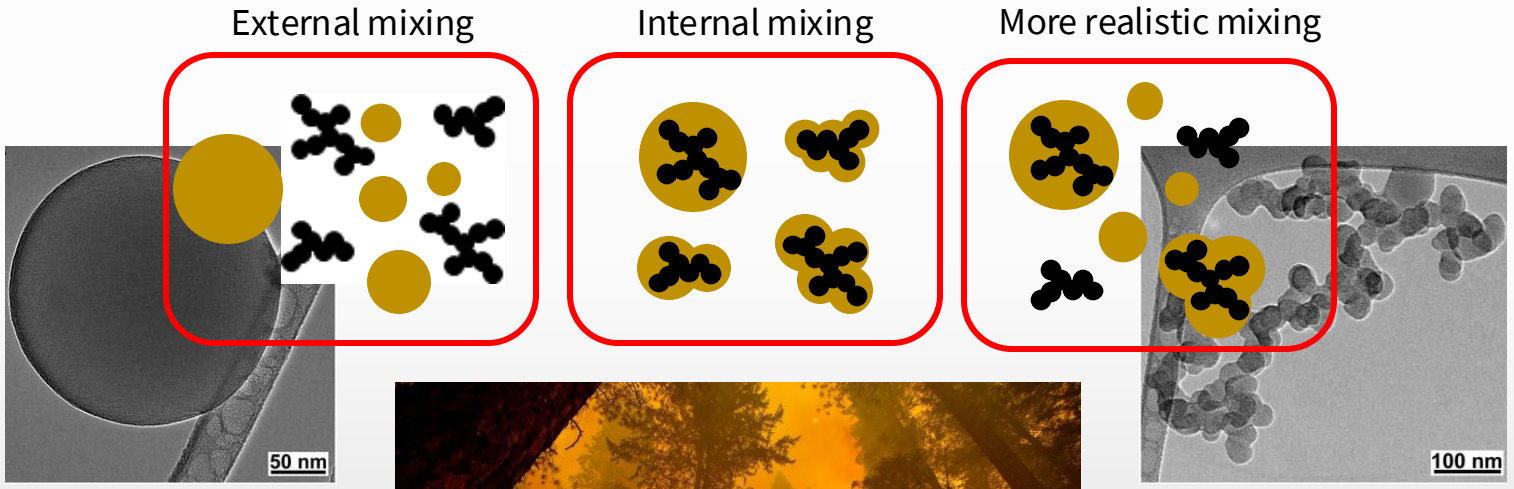
- Organic carbon (OC):
  - Primarily emitted during pyrolysis and smoldering
  - Wide range of refractory properties
  - Brown carbon: subset of OC absorbs strongly at lower visible and ultraviolet wavelengths



Pöschl, 2003, Anal. Bioanal. Chem.

Aerosols from Wildfires

- Biomass burning emissions are one of the largest sources of organic aerosol

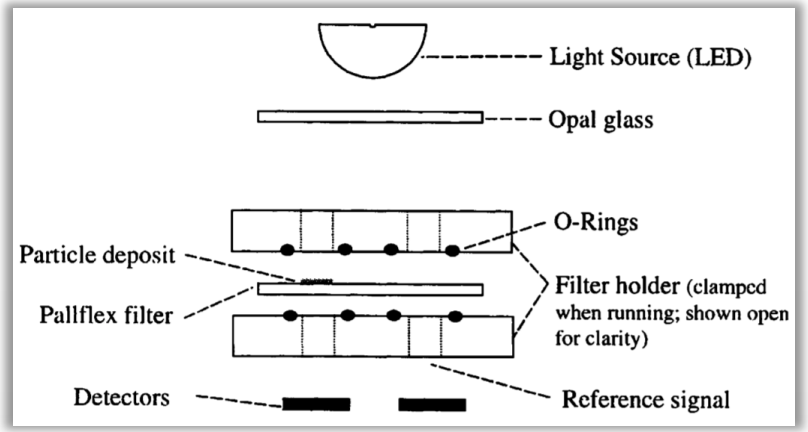


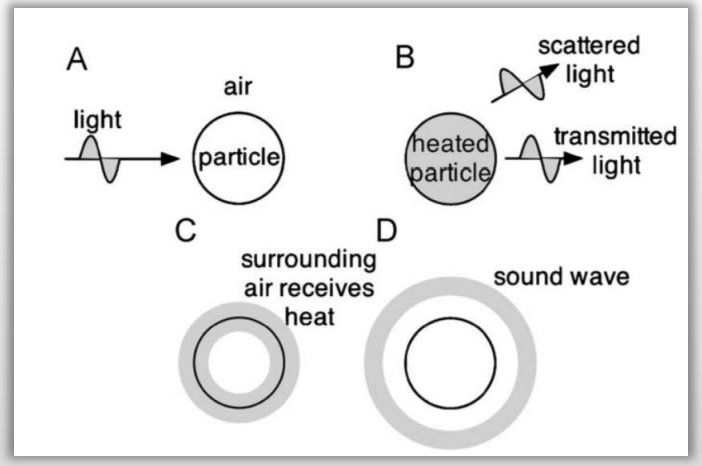
- Mixing state of OC and BC m… their optical properties
  - **Difficult to isolate** OC and B…
  - **OC optical properties chang**…g

Alexander et al., 2008, Science

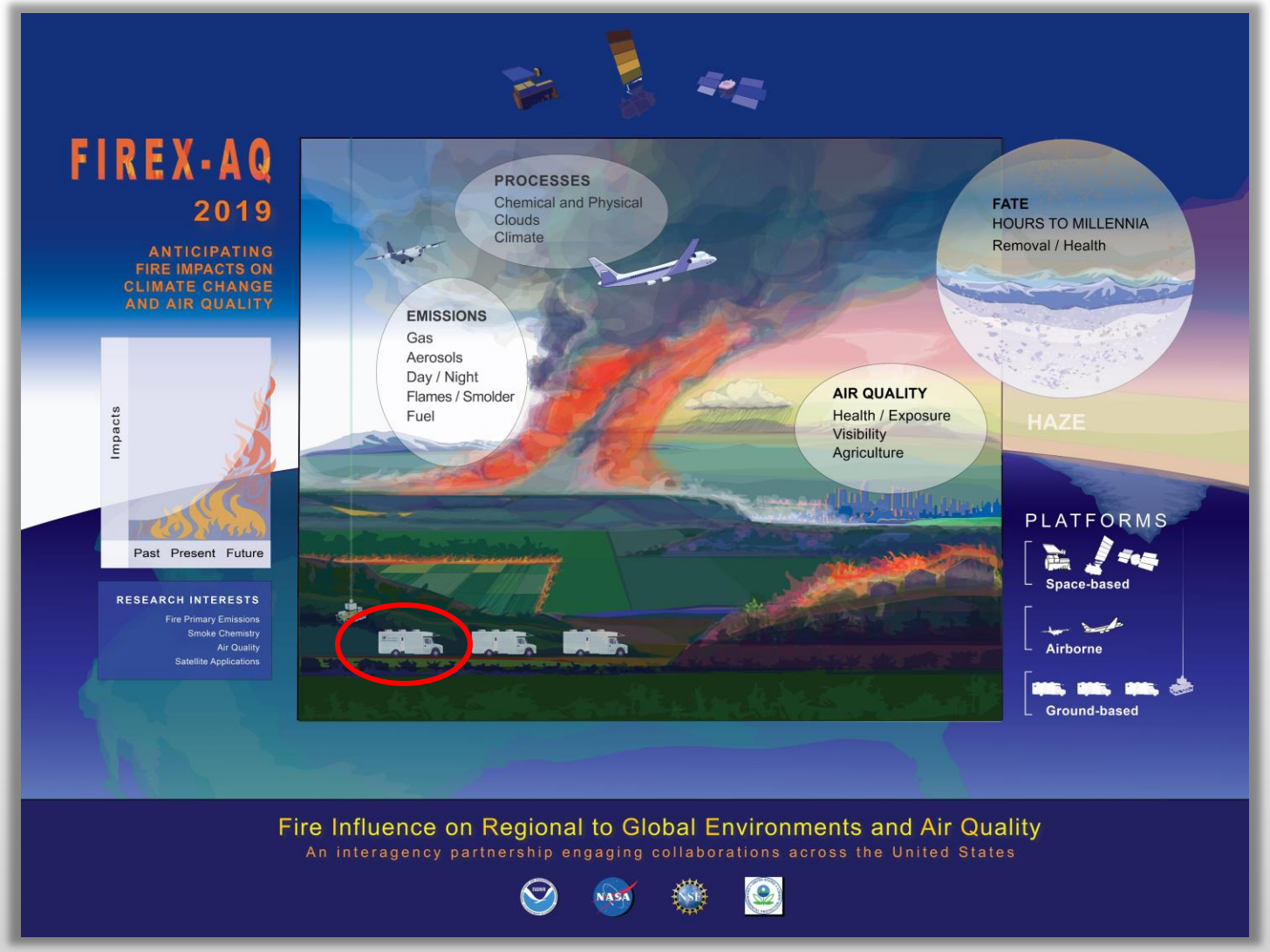
Optical Property Measurements

- Filter-based – Aethalometer, Particle Soot Absorption Photometer, Tricolor Absorption Photometer
- In-situ – Photoacoustic spectroscopy, difference between Extinction and Scattering (nephelometer)
- Electron Energy Loss Spectroscopy (EELS)





Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)



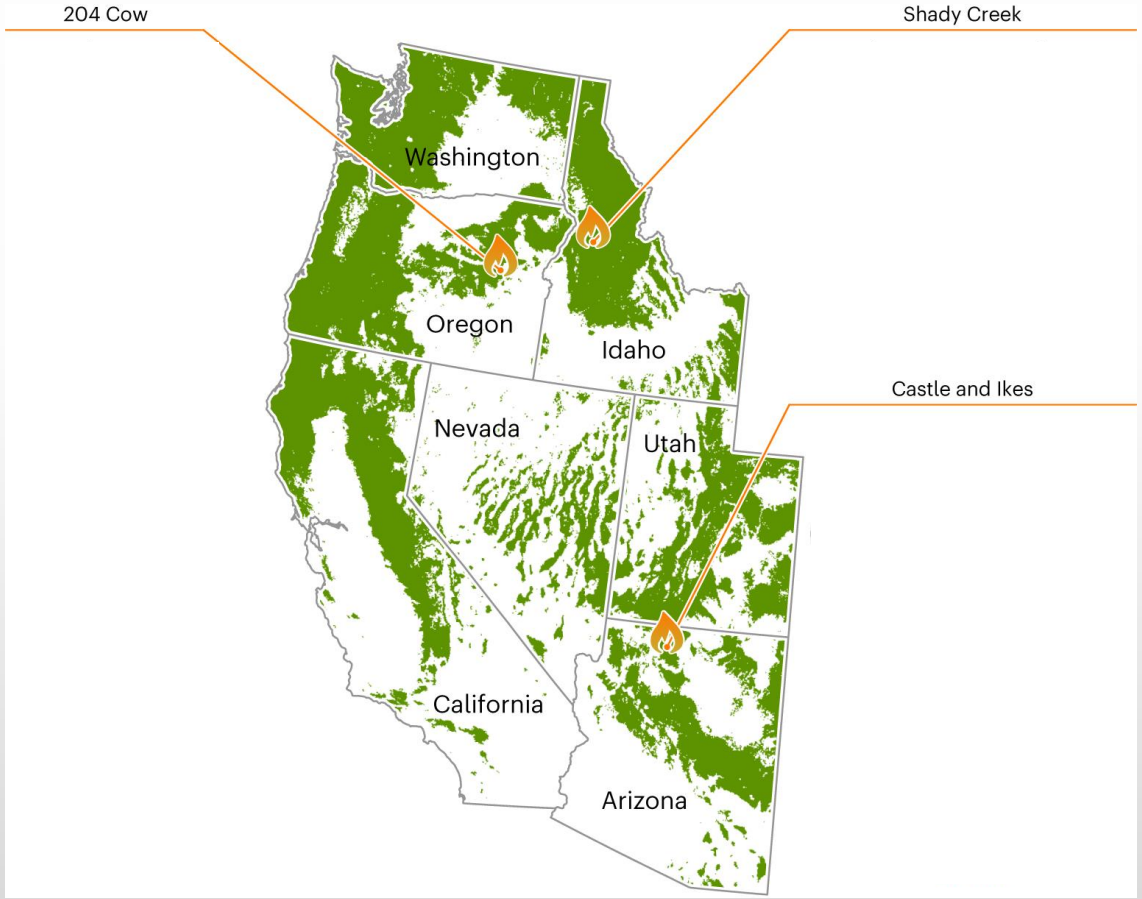
Sampled Fires Across the Western U.S.

Research Questions

1. What factors control the optical properties of organic aerosols from wildfires?

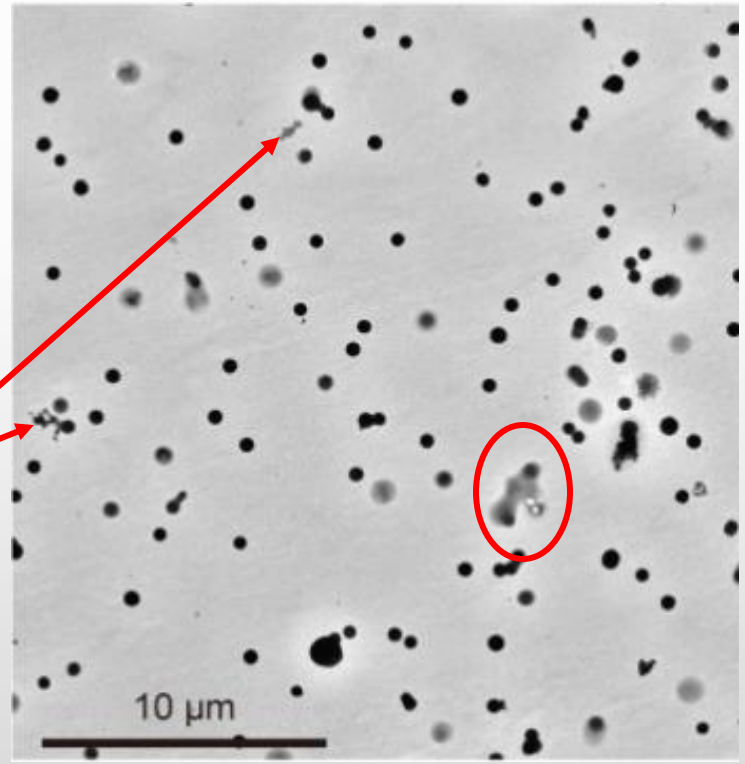
2. What is the dependence of these properties on BrC chemical composition?

Aerosol Light Absorption at Sampling Locations

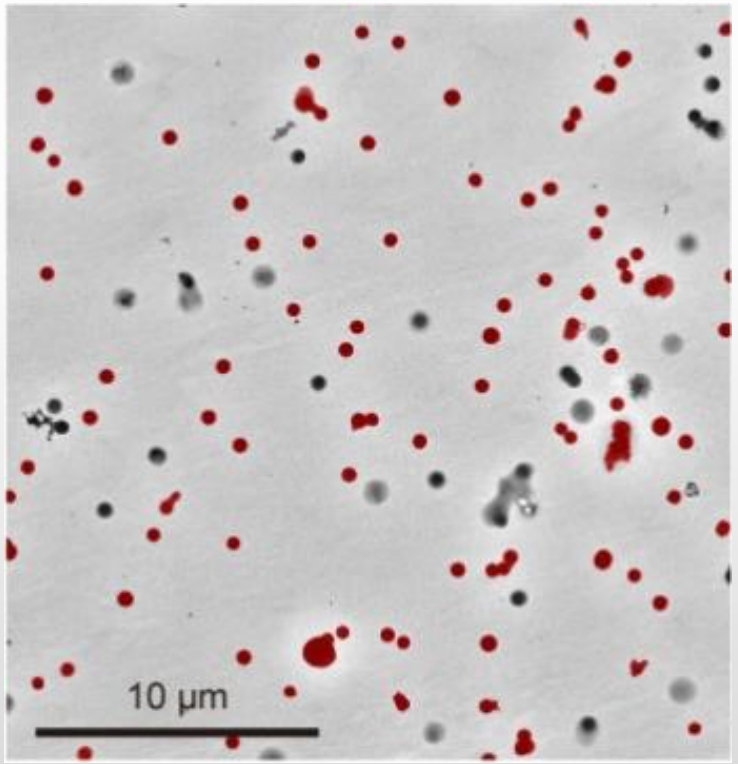


Dark Particles on Microscope Grids

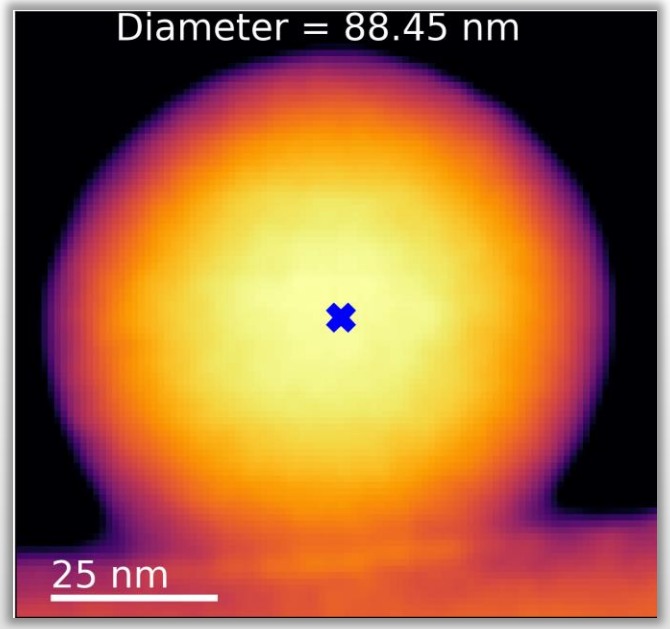
All particles



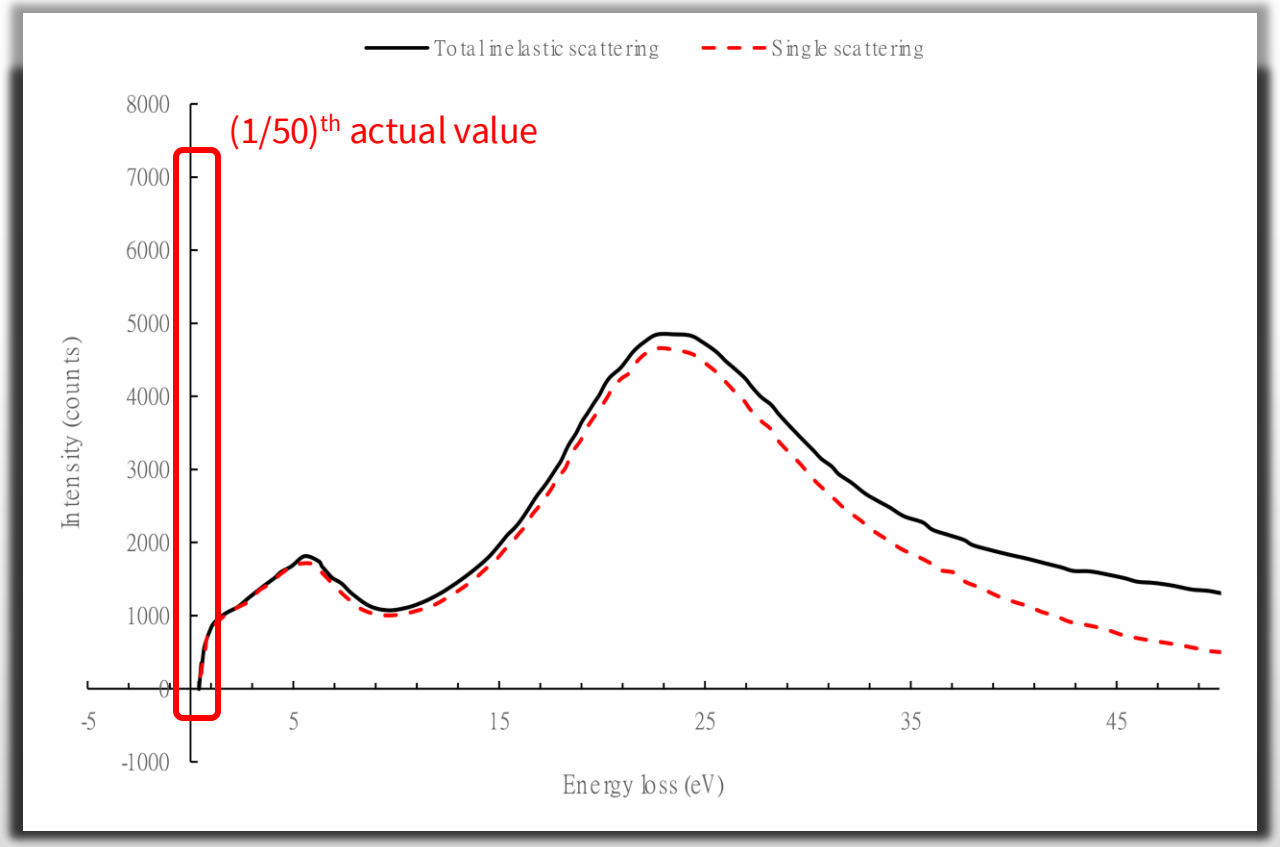
"Dark" particles in red



Electron Energy Loss Spectroscopy (EELS) of “dark” BrC



Performed at Oakridge National Lab

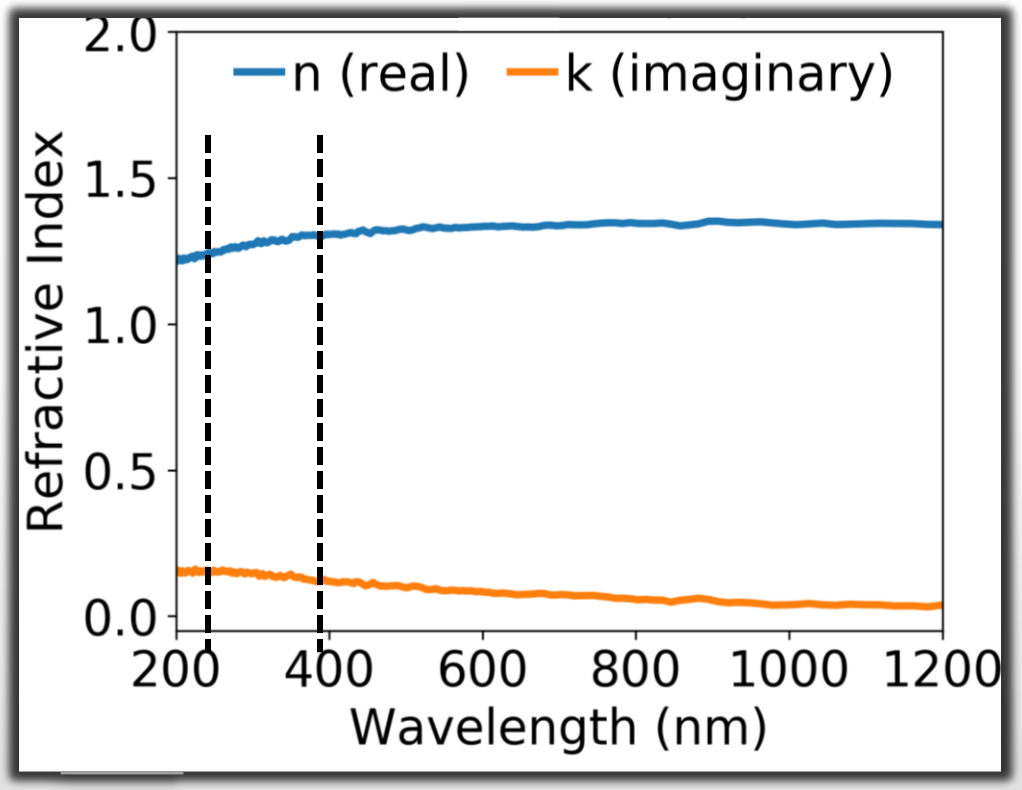


Refractive Index from Single Scattering Signal

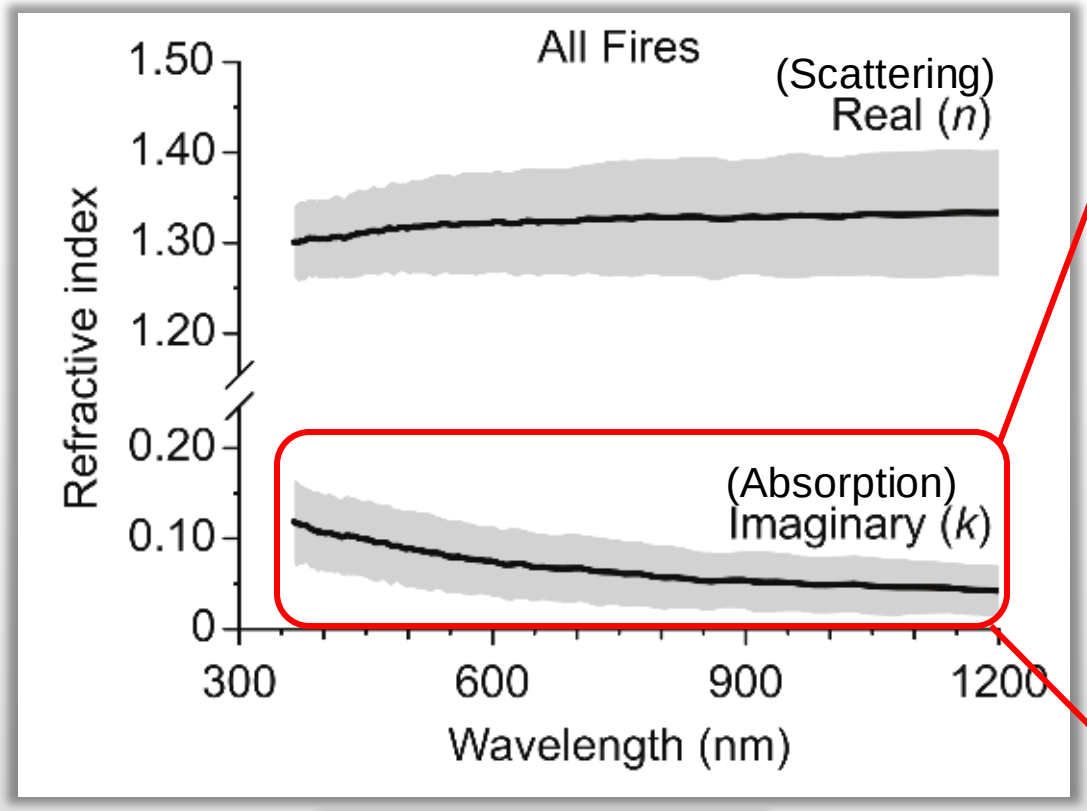
$$S(E) = \frac{I_0 t}{\pi a_0 m_0 v^2} \mathrm{Im}\left[\frac{-1}{\varepsilon(E)}\right] \ln\left[1 + \left(\frac{\beta}{\theta_E}\right)^2\right] + S_S(E),$$

$$\mathrm{Re}\left[\frac{1}{\varepsilon(E)}\right] = 1 - \frac{2}{\pi} P \int_0^\infty \mathrm{Im}\left[\frac{-1}{\varepsilon(E')}\right] \frac{E' dE'}{E'^2 - E^2}$$

$$\varepsilon(E) = \varepsilon_1(E) + i\varepsilon_2(E) = \frac{\mathrm{Re}\left[1/\varepsilon(E)\right] + i\,\mathrm{Im}\left[-1/\varepsilon(E)\right]}{\left\{\mathrm{Re}\left[1/\varepsilon(E)\right]\right\}^2 + \left\{\mathrm{Im}\left[-1/\varepsilon(E)\right]\right\}^2}$$

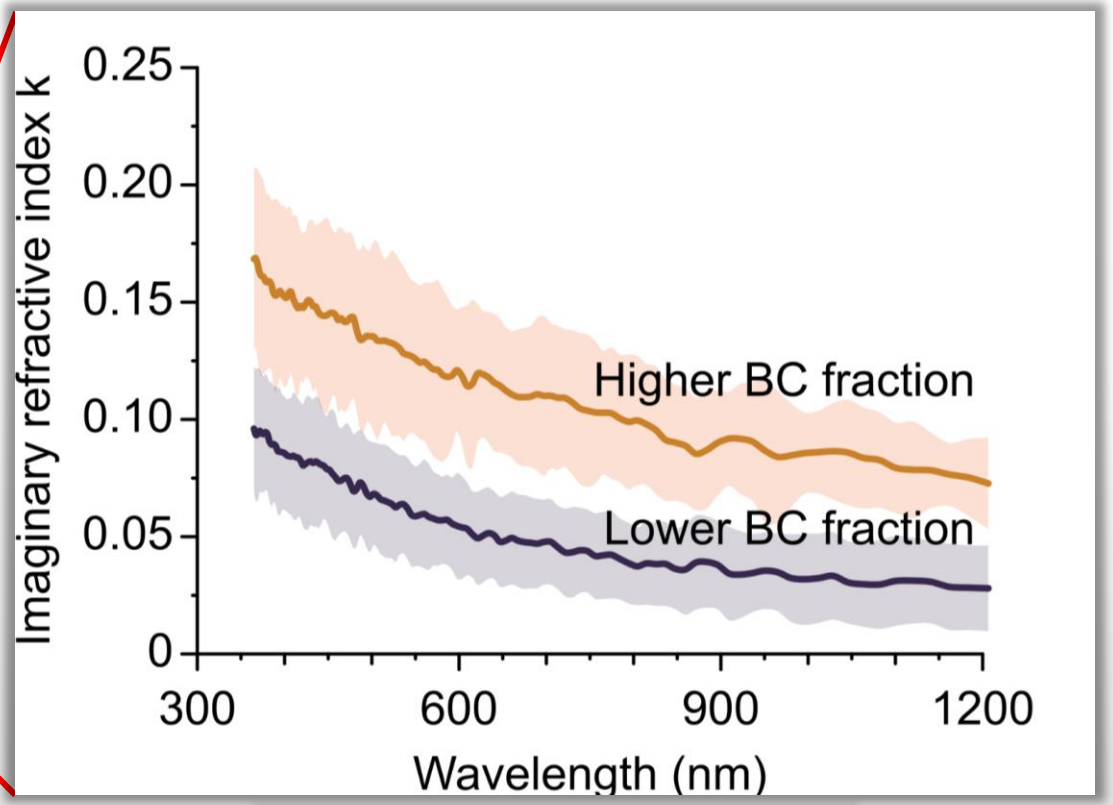


Spectral Refractive Index of dark BrC



$k_{all\ fires} = 9.7(\lambda^{-0.762})$

λ is in nm

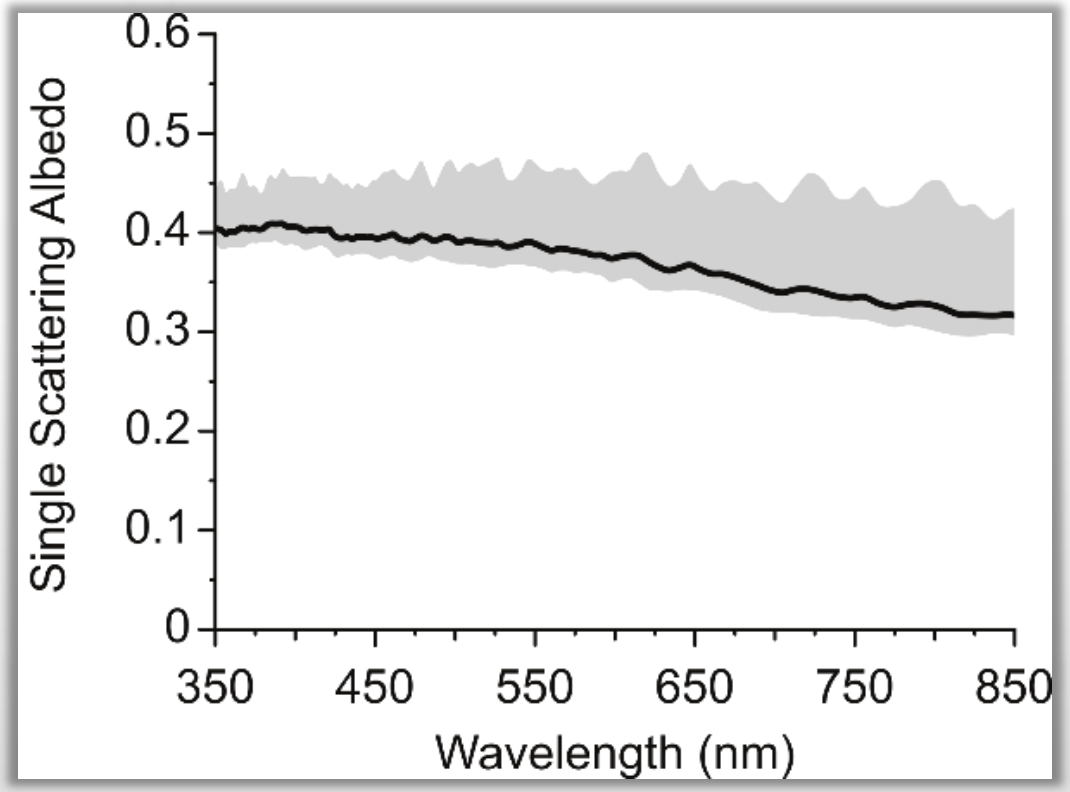


$k_{Higher\ BC\ fraction} = 7.9(\lambda^{-0.65})$

$k_{Lower\ BC\ fraction} = 57(\lambda^{-1.08})$

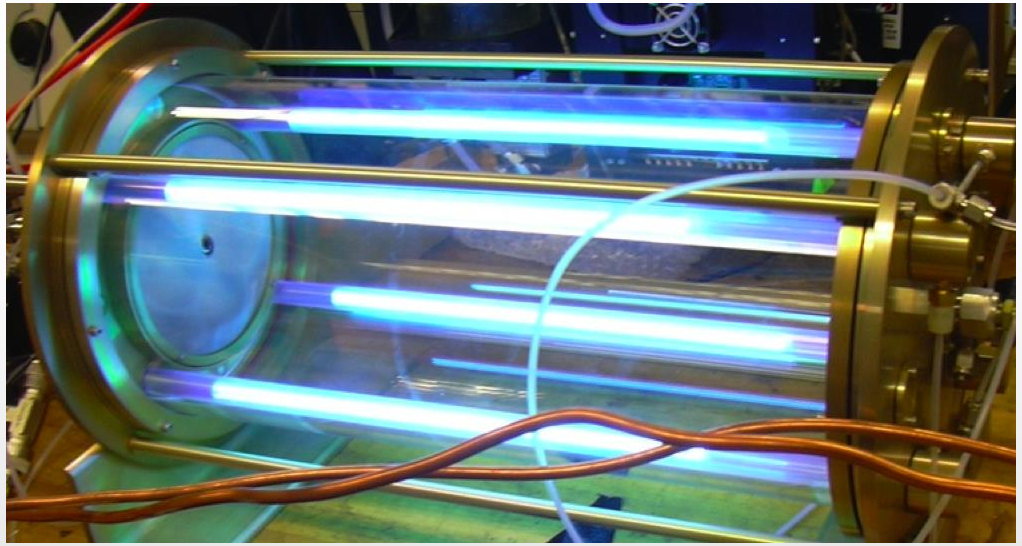
Optical Properties

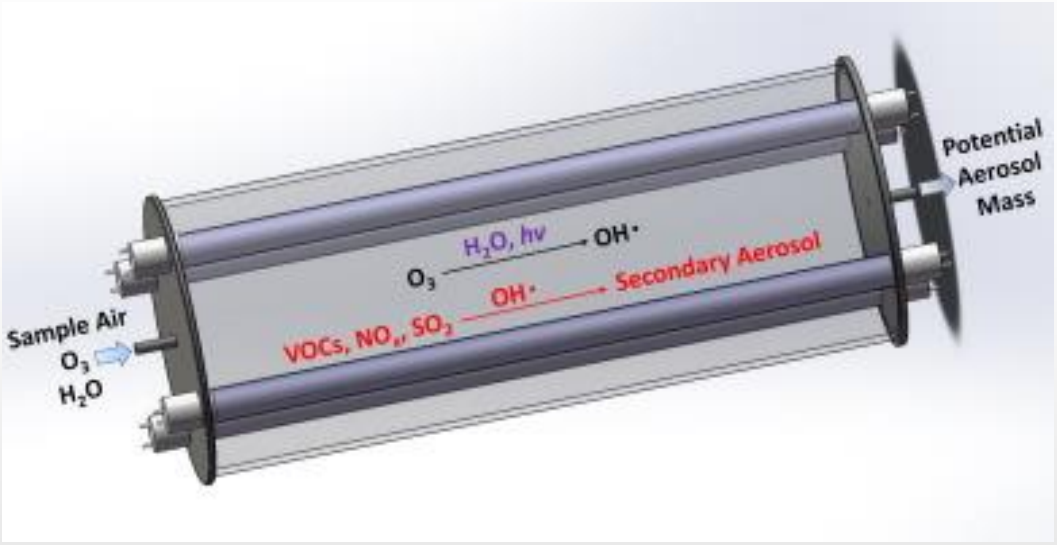
- The single scattering albedo (ratio of scattering coefficient to extinction coefficient) for these particles was 0.38 ± 0.03.
  - For comparison soot has SSA between 0.1 and 0.3.



Reactor to Simulate Atmospheric Processing

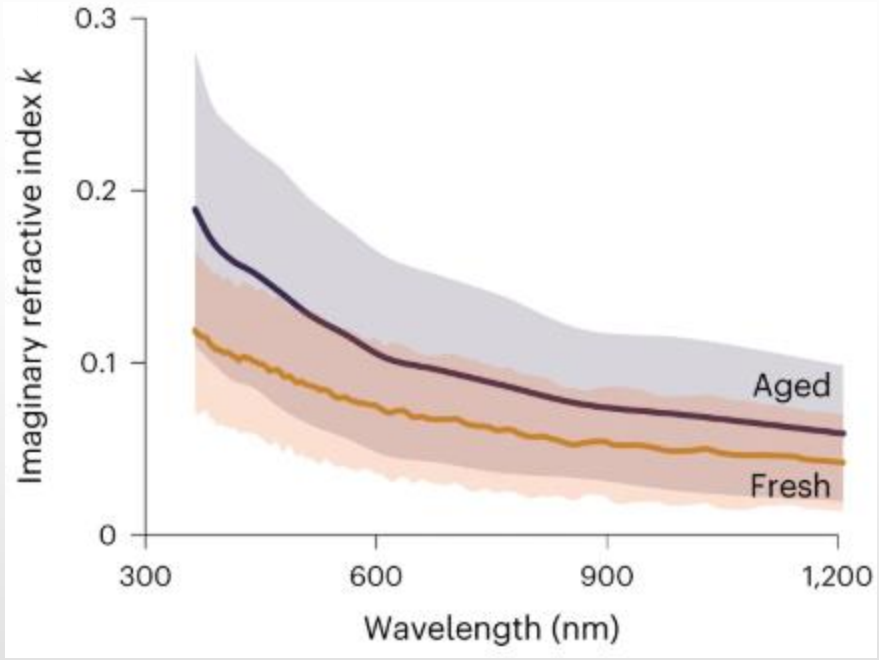
Potential Aerosol Mass (PAM) reactor



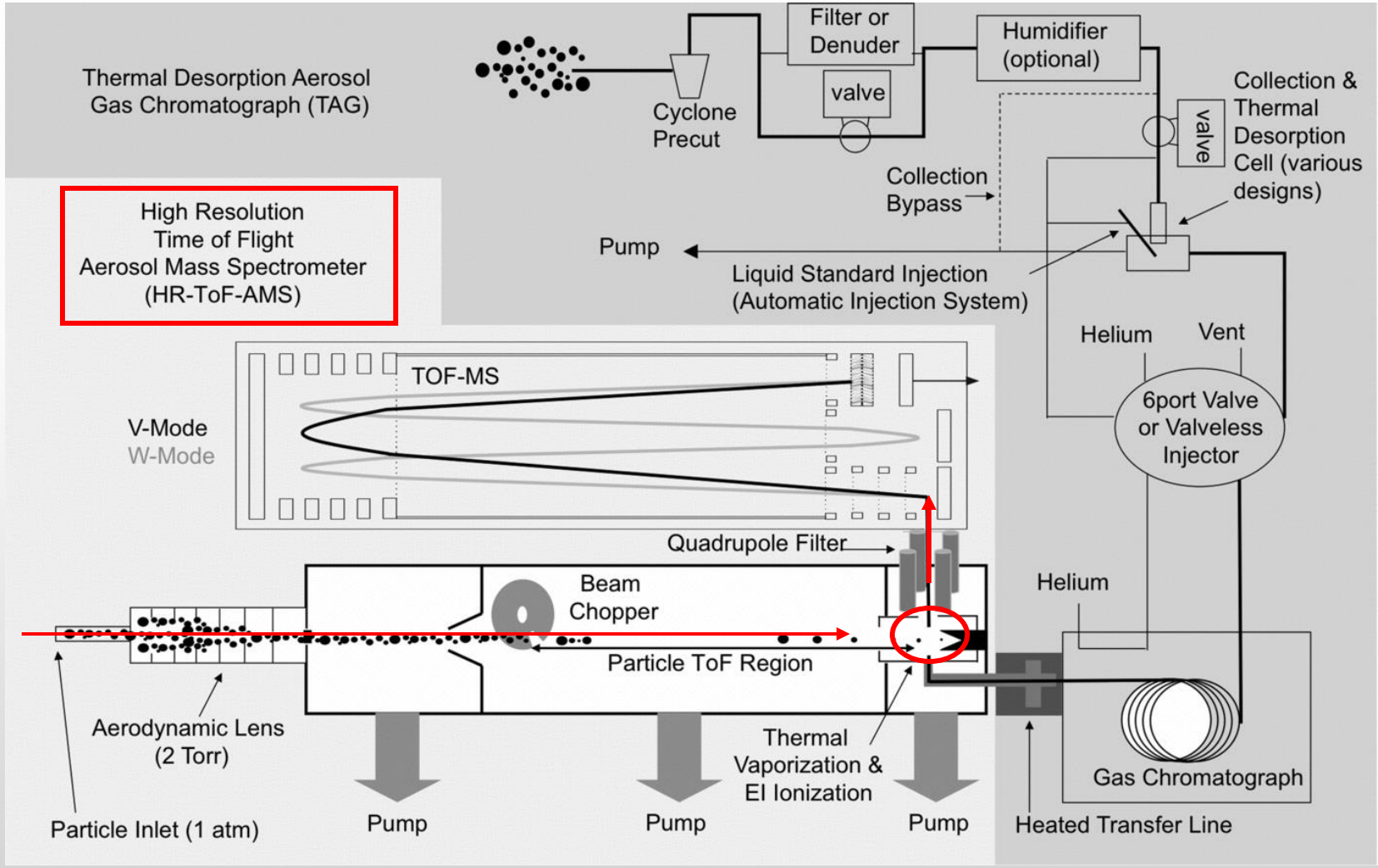


Simulated Aging Increased Light Absorption

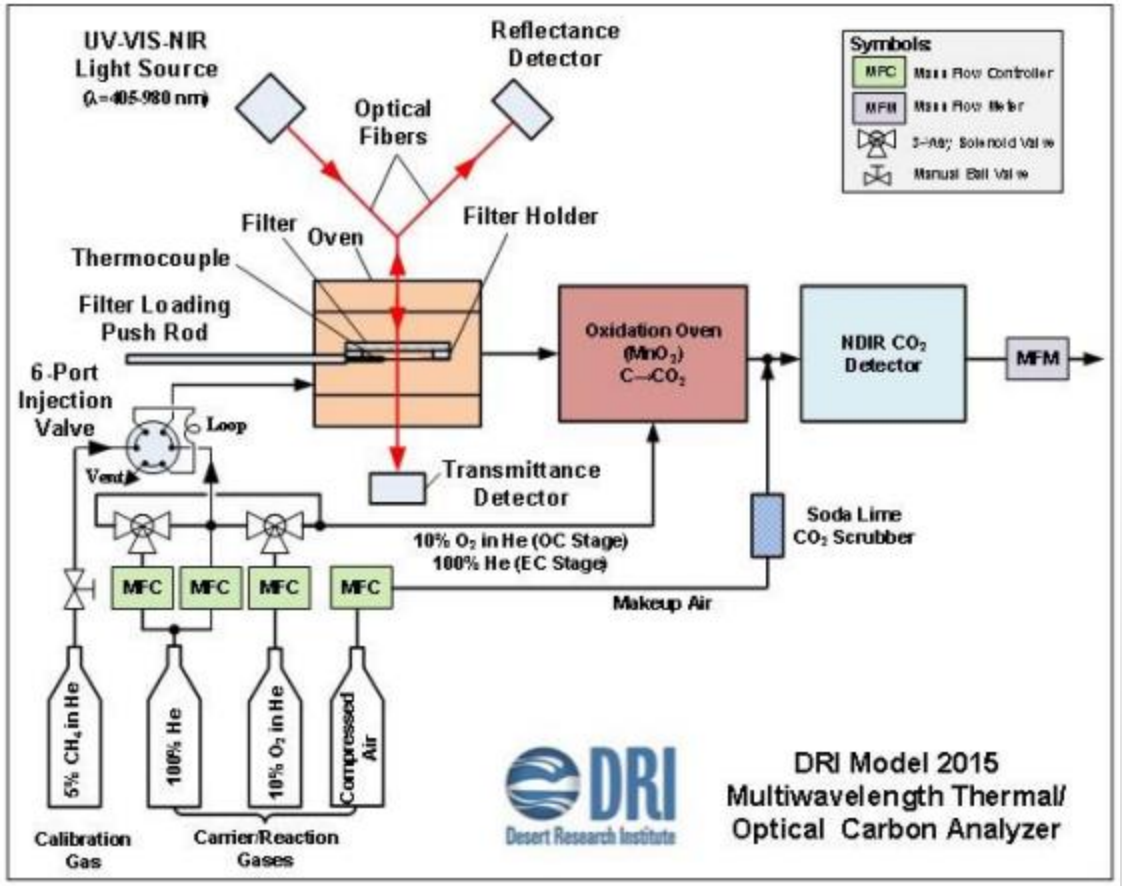
- Processing through a PAM reactor resulted in increased light absorption by the particles.
- Propagated errors were high due to uncertainties in PAM measurements.

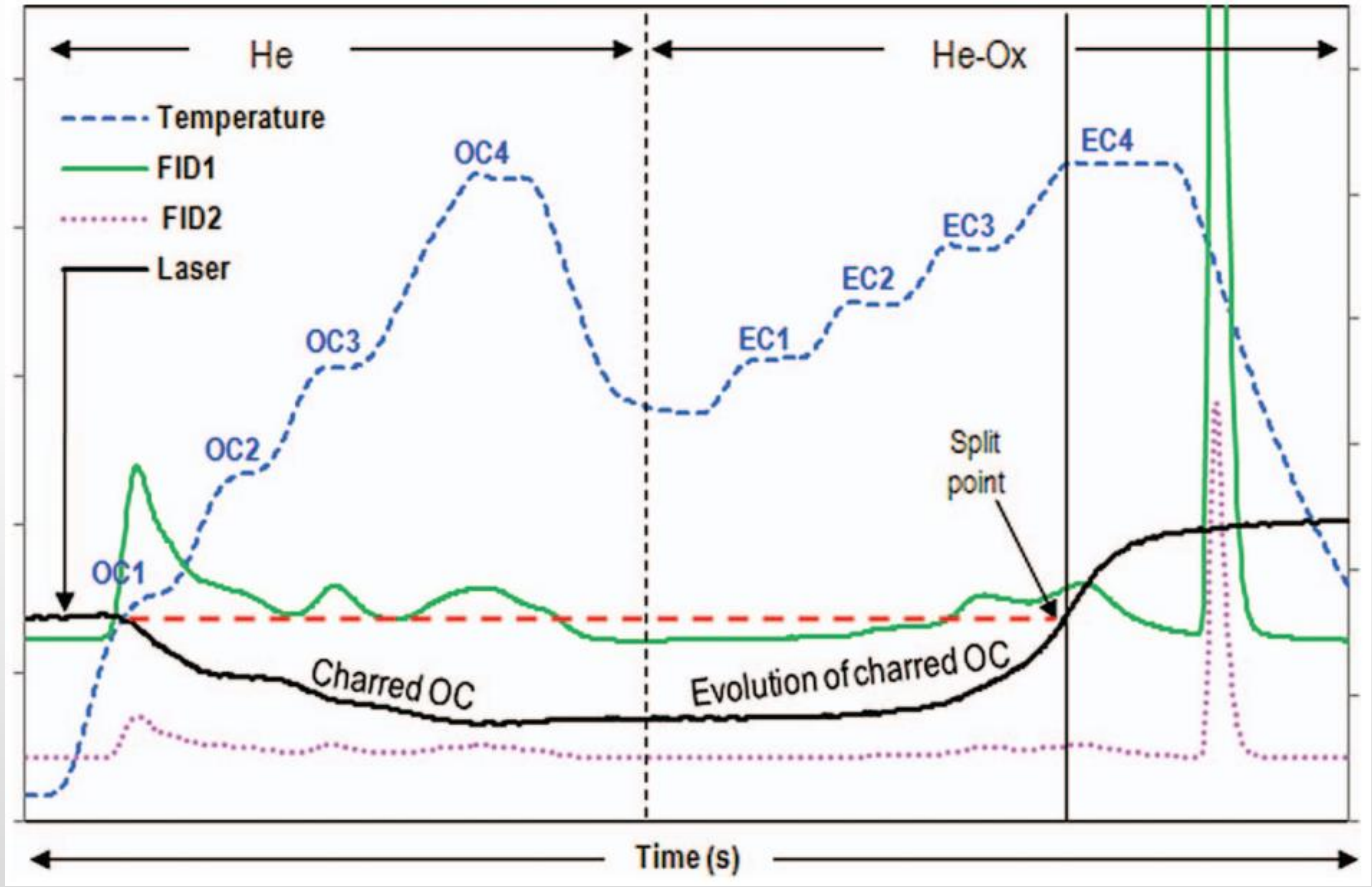


Chemical Composition Measurement

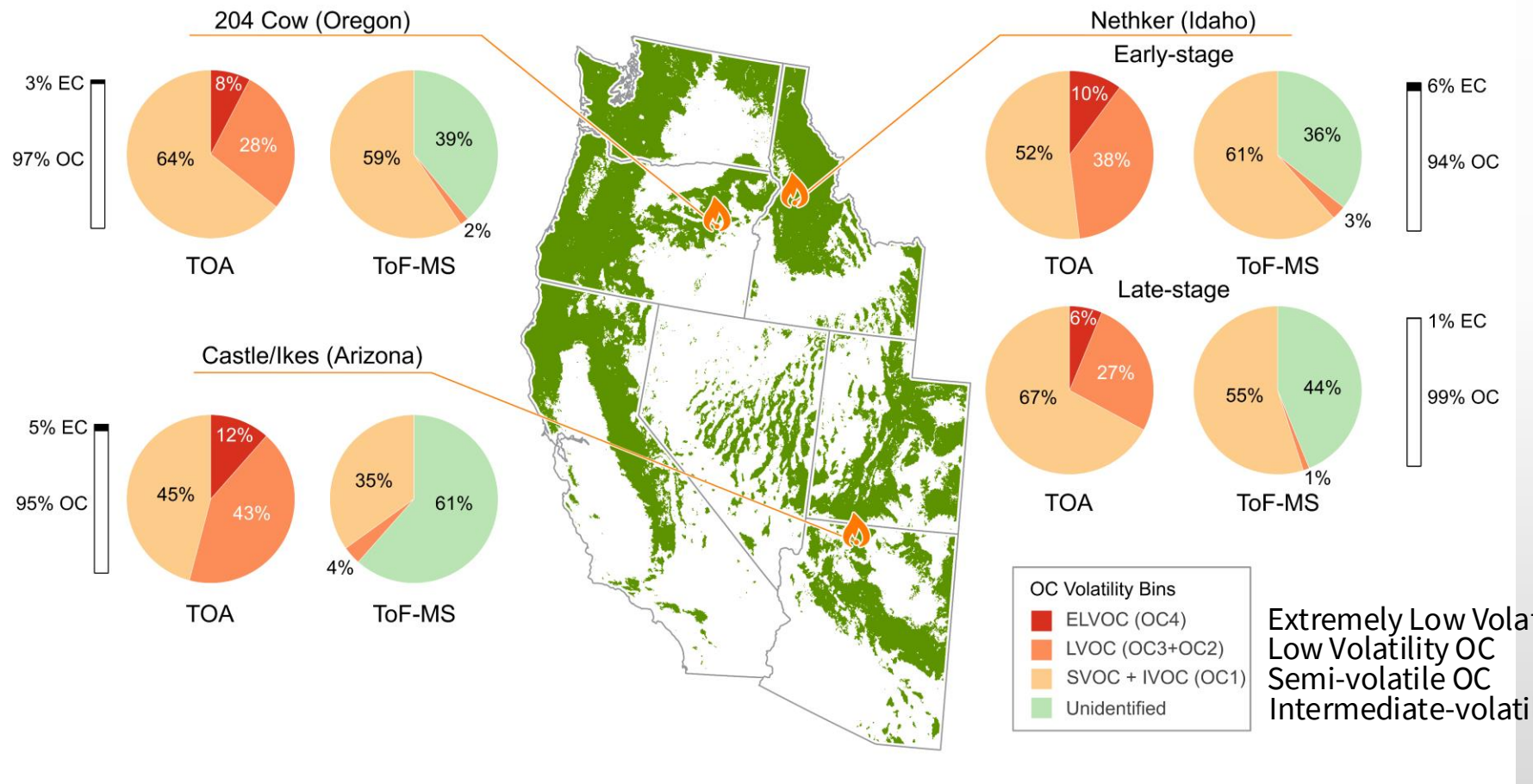


Composition Measurement—Thermal-Optical Analysis





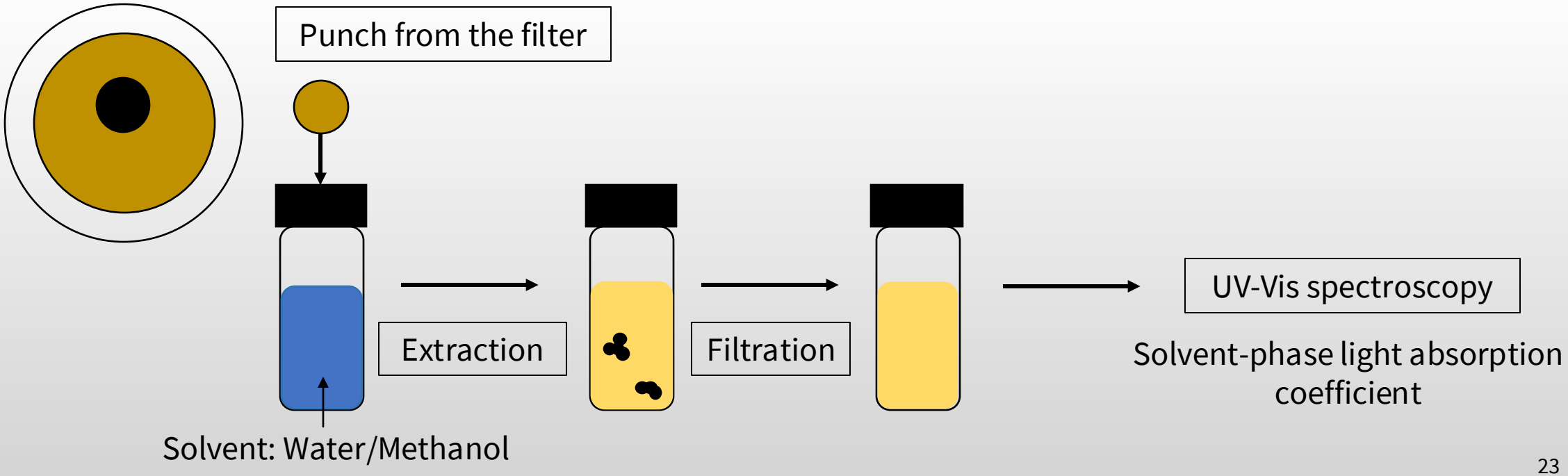
Chemical Composition of Smoke



Extremely Low Volatility OC
Low Volatility OC
Semi-volatile OC
Intermediate-volatility OC

Isolating OA light absorption

- Measuring organic aerosol (OA) absorption:
  - Collect aerosols on a substrate
  - Extract organics into a solvent
  - Measure light absorption by dissolved BrC molecules (chromophores)



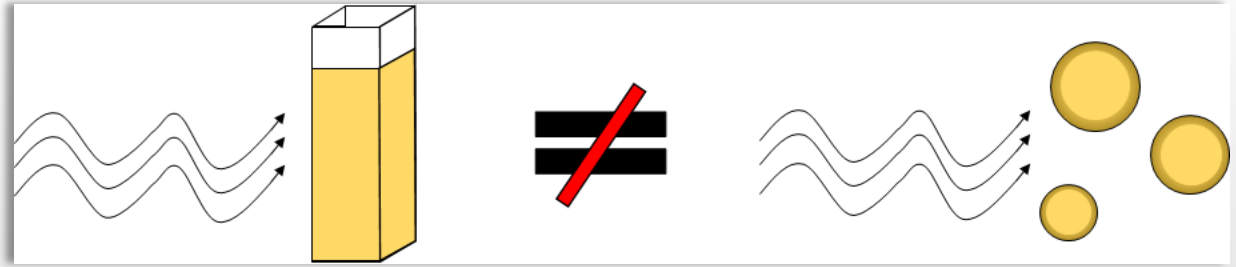
Shetty et al., 2019, Atmospheric Chem. Phys.

Difference in Particle and Solvent Light Absorption

- Absorption coefficient by OA particles ($b_{abs,OA}$) is:

$$b_{abs,OA} = \int \sigma_{abs} \cdot n_d(d_p) \cdot d(d_p)$$

The absorption cross section (σ_{abs}) has units of m^2 while the size distribution function ($n_d(d_p)$) has units of $\#/(cm^3.nm)$. So, $b_{abs,OA}$ has units of $Length^{-1}$ (order of Mm^{-1})



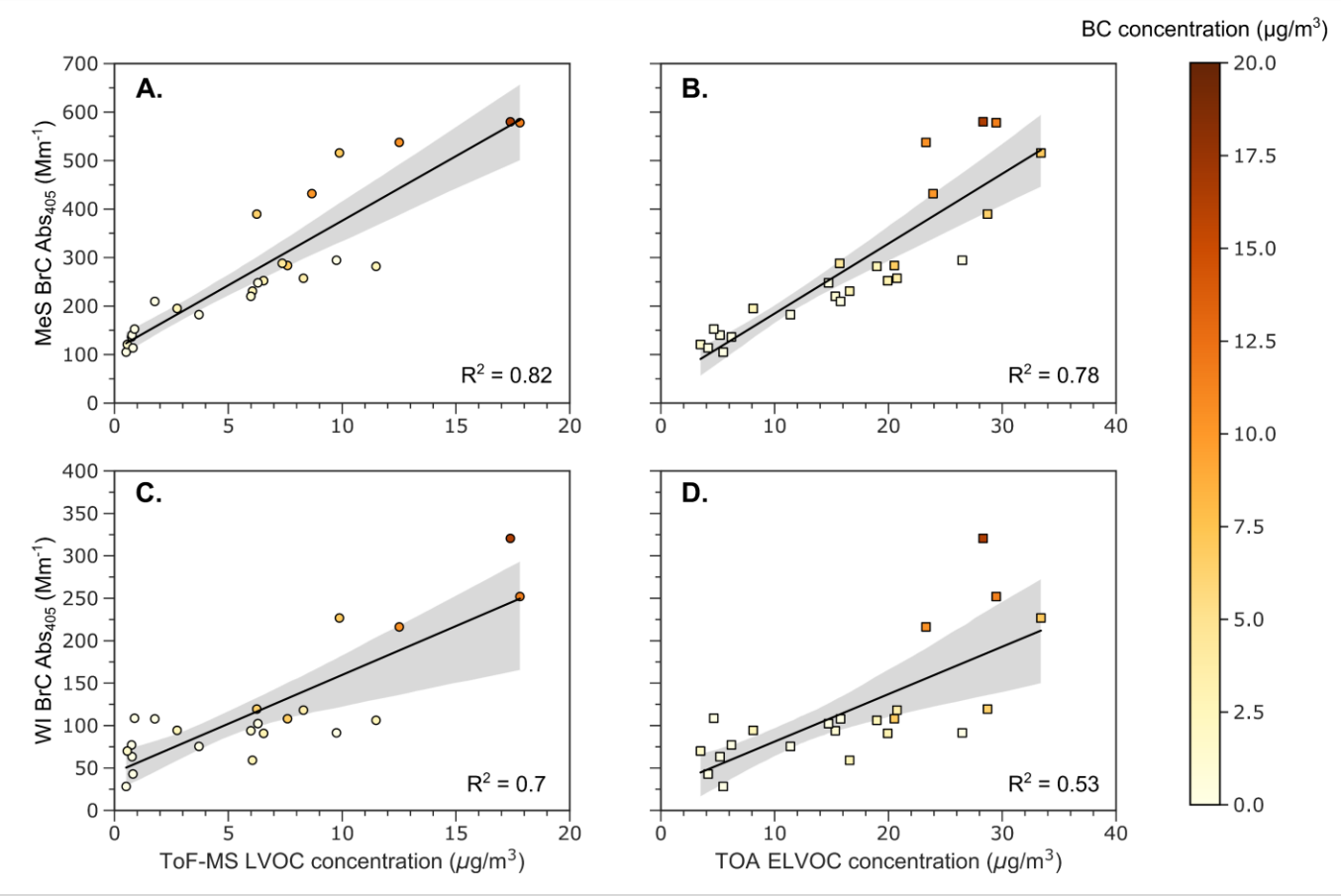
- A factor of 2 was typically used for corrections.
- I previously showed that the factor may severely underestimate light absorption by OA

Washenfelder et al., 2015, *Geophys. Res. Lett.*
Shetty et al., 2019, Atmospheric Chem. Phys.

Light-absorption by Soluble Brown Carbon (BrC)

- The absorption coefficient by both the methanol-soluble (MeS) and water-insoluble (WI) fraction of BrC were positively associated with low-volatility organic carbon (LVOC) concentrations.

- Samples with higher BC concentrations had greater light absorption at similar Extremely-low/LVOC concentrations.
  - Likely that higher temperatures that lead to more BC formation are also conducive to forming BrC with greater light absorption.

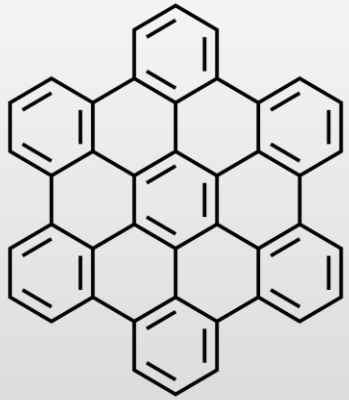


ToF-MS – Time of Flight Mass Spectrometry

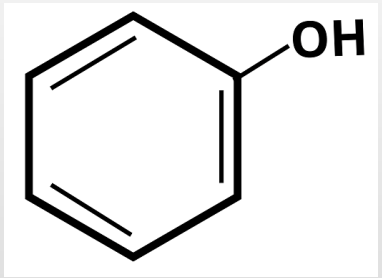
TOA – Thermal Optical Analysis

Most Absorbing Compound Groups

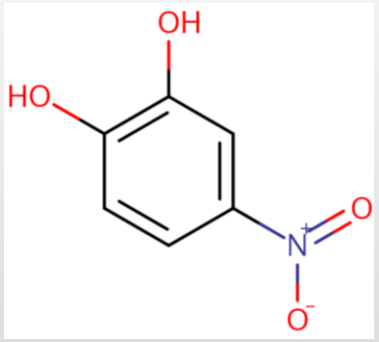
- Through a multi-variate regression, we found the most light-absorbing compounds groups were:
  - Polycyclic Aromatic Hydrocarbons (PAHs),
  - Aromatics, and
  - Nitrogen-containing Organics (NOCs)



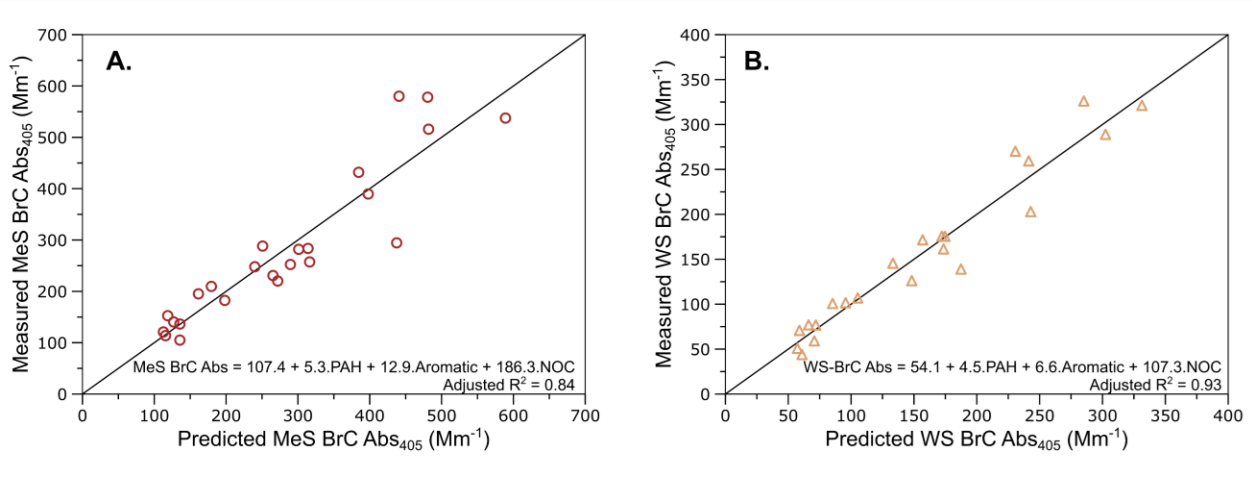
PAHs



Aromatic

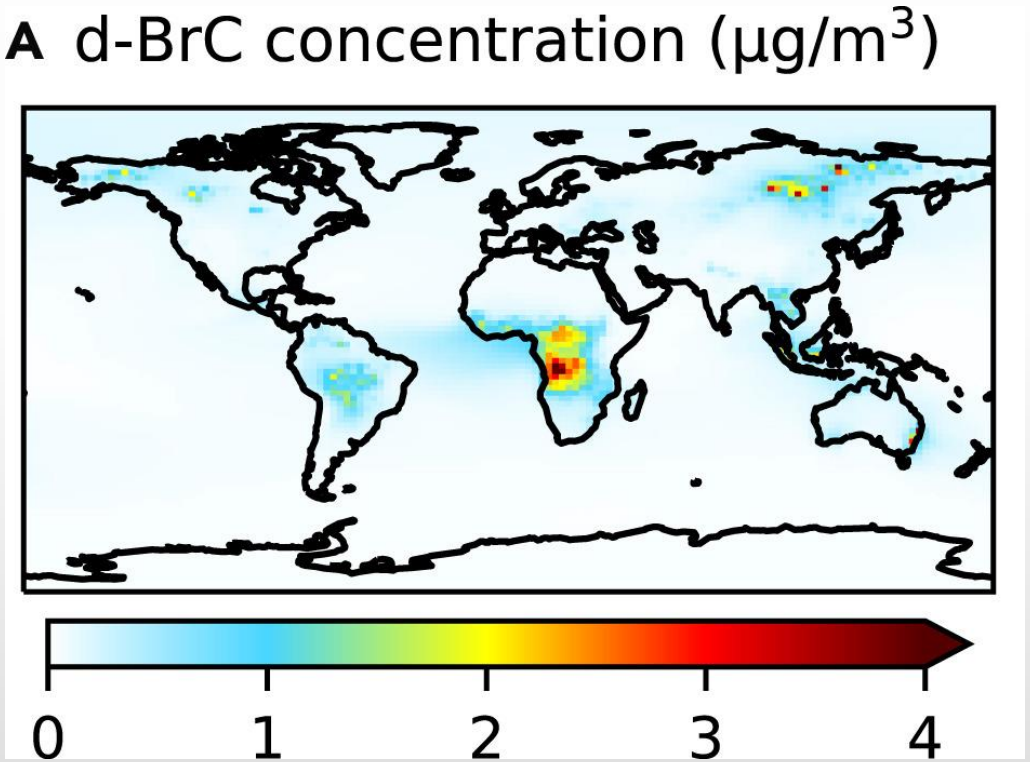


NOCs



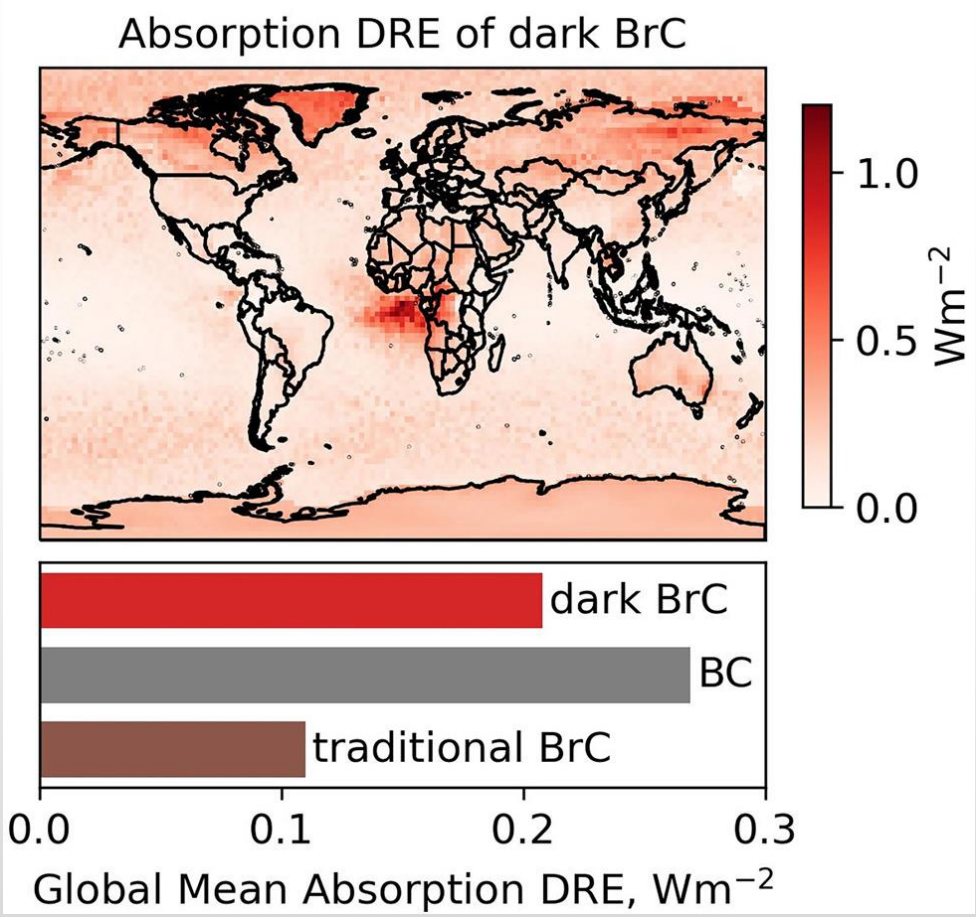
D-BrC Across the Globe

- Models based on emission inventories and satellite measurements are predicting presence of d-BrC across the globe.
- Values seem low since these are annual averages, and d-BrC generating events are transient.
- Predictions over Africa, Siberia, and Southeast Asia were ~4 µg/m^3 which is around 20% of total OA from biomass burning.



Global Implications

- Including d-BrC into radiative calculations reveals a net warming by organics.
- The warming is comparable to that of Black Carbon.
- These values are likely biased low since emission inventories used in the model are conservative.



Key Takeaways

1. What factors control the optical properties of organic aerosols from wildfires?
   - The d-BrC emissions **absorbed light even at longer wavelengths** and the **absorption increased with BC fractions.**
   - The **soluble brown carbon** concentrations were **associated with BC** indicating that they may be co-emitted in fires.
2. What is the dependence of these properties on BrC chemical composition?
   - **PAHs**, **Aromatic compounds**, and **Nitrogen-containing organics** were the dominant constituents of BrC.
   - Lower volatility organics are efficient light-absorbers.

Shetty, Nishit, et al. *Environmental Science: Atmospheres* 3.9 (2023): 1262-1271. (*Highlighted on journal cover*)
Chakrabarty, Rajan K., Shetty, Nishit, et al. *Nature Geoscience* (2023): 1-6.

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