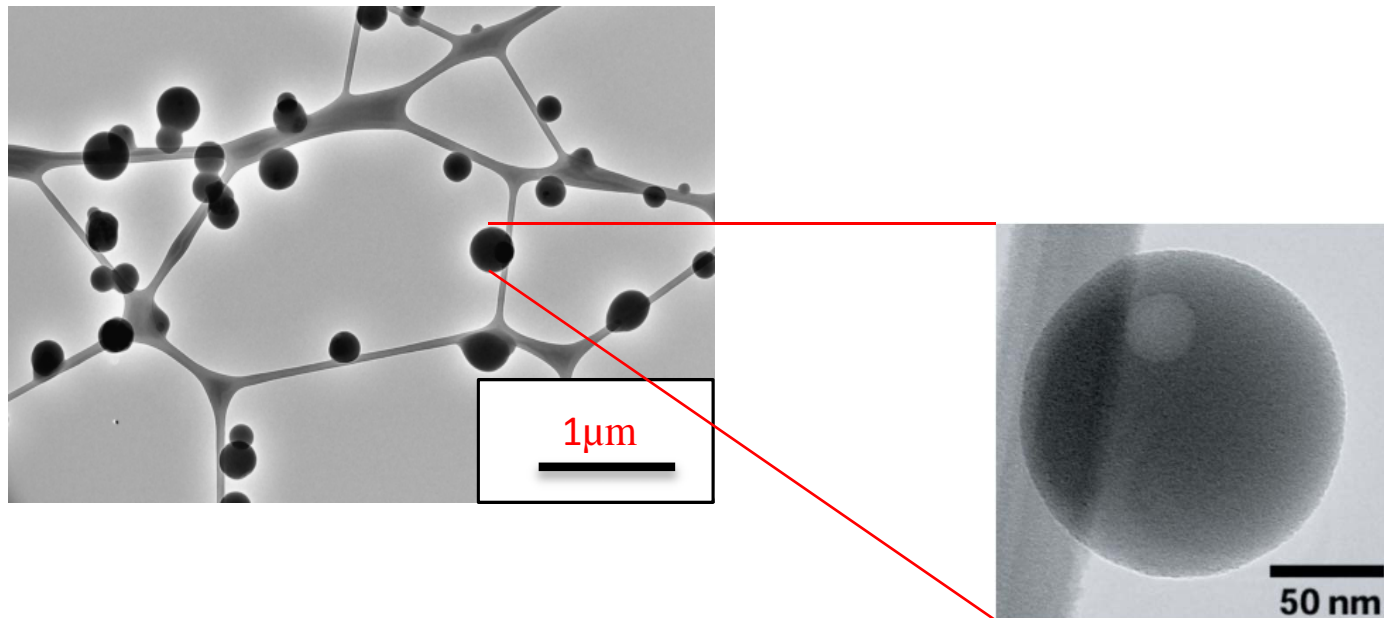


Tar Balls Observed in Wildfire Plumes are Weakly Absorbing Secondary Aerosols

A. J. Sedlacek, P. Buseck, K. Adachi, L. Kleinman, T. Onasch, and S. R. Springston



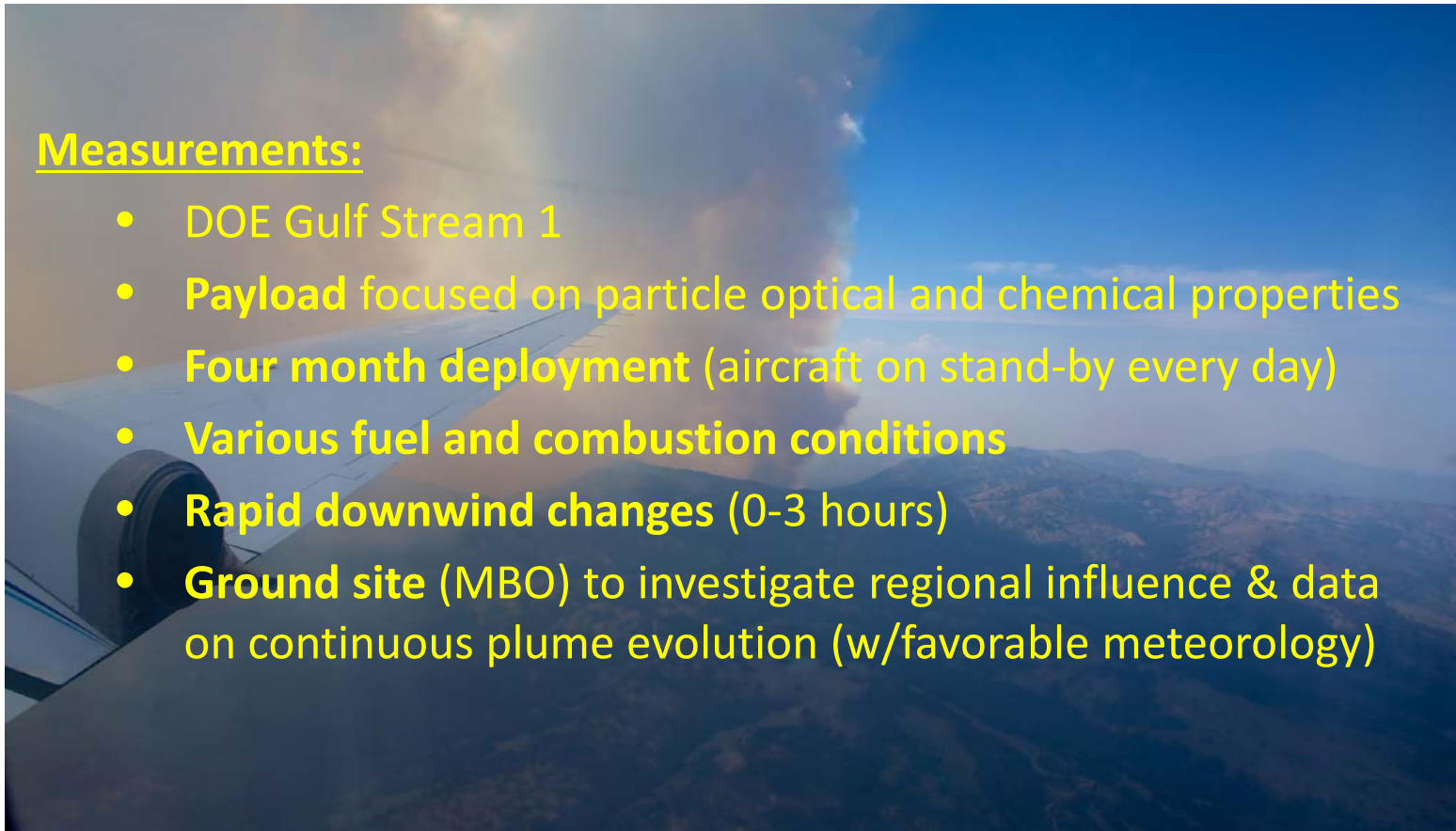
DOE-Sponsored *Biomass Burn Observation Project (BBOP)*

Scientific Motivation:

To understand and quantify the role of BB aerosols in climate forcing by investigating the near field evolution of their chemical, hygroscopic, microphysical, and optical properties

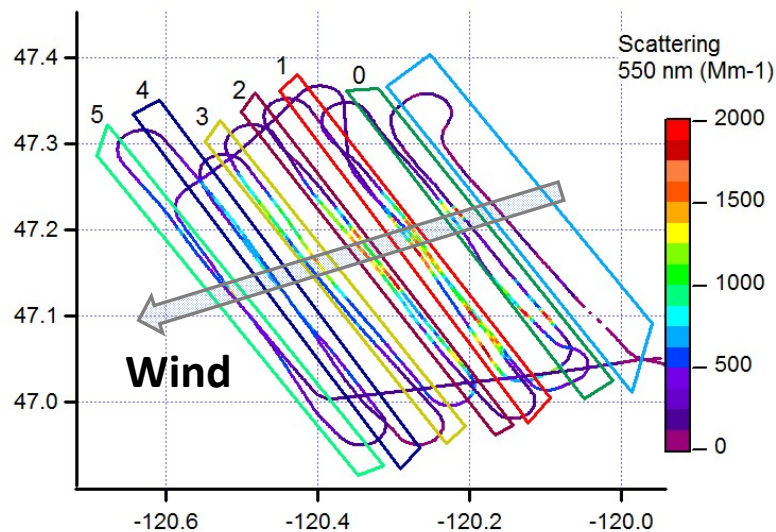
Measurements:

- DOE Gulf Stream 1
- **Payload** focused on particle optical and chemical properties
- **Four month deployment** (aircraft on stand-by every day)
- **Various fuel and combustion conditions**
- **Rapid downwind changes** (0-3 hours)
- **Ground site** (MBO) to investigate regional influence & data on continuous plume evolution (w/favorable meteorology)

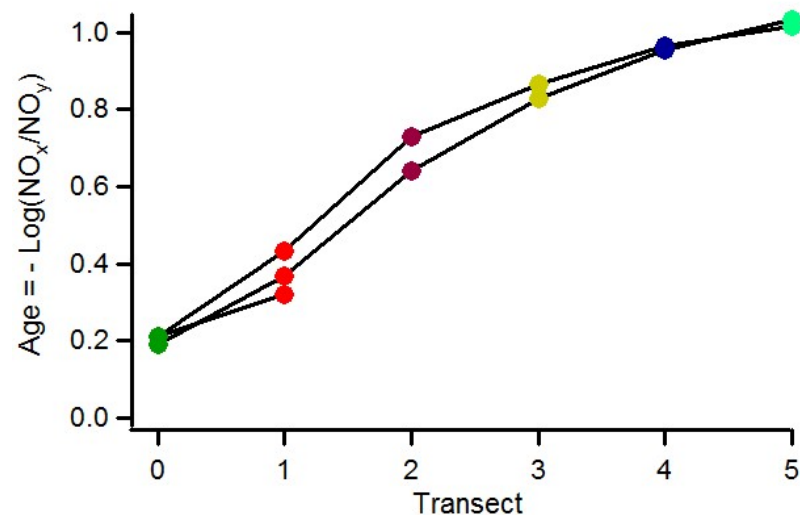


Lagrangian Flight Pattern

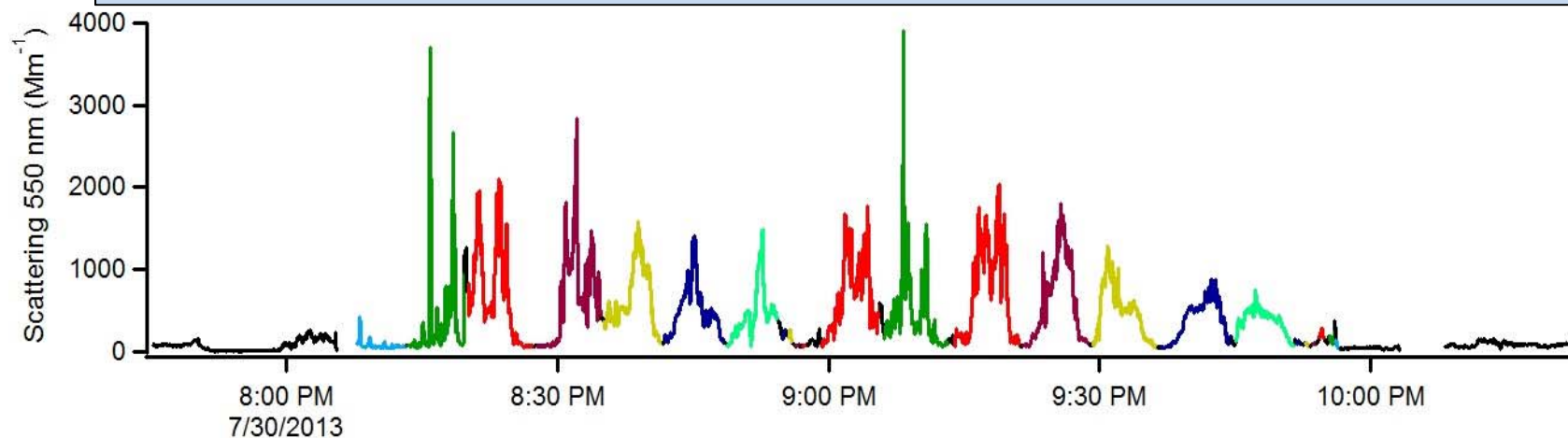
Colockum Tarps Fire, Flight 730b



Photochemical Age increases downwind

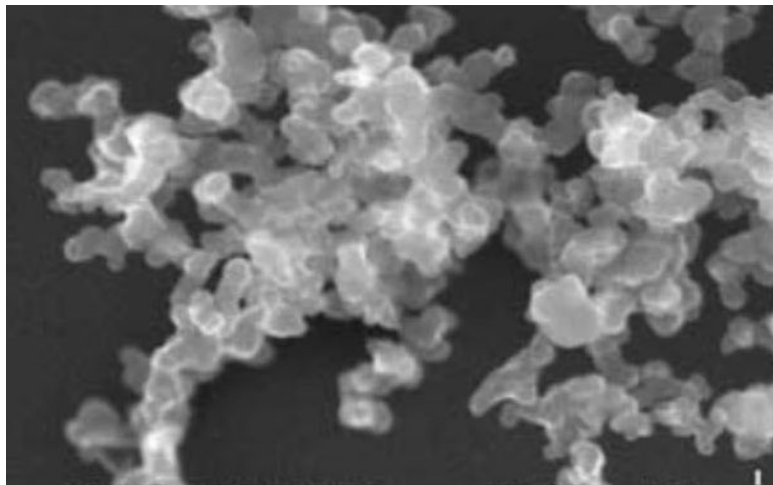


Two sets of 6 transects ~ 0 to 2.5 hours aging. Plume dilutes downwind



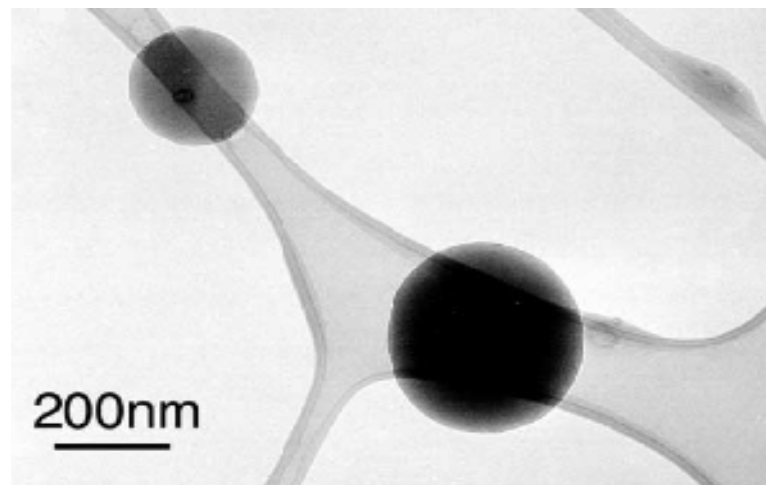
Types of Spherical Carbonaceous Solids

Soot



Li, Pósfai, Hobbs, Buseck (JGR 2003)

Tar balls (BrC particles)



Li, Pósfai, Hobbs, Buseck (JGR 2003)

Tar Balls (TBs)

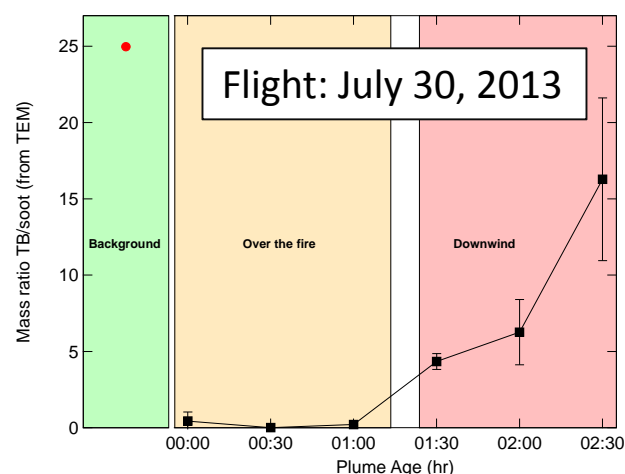
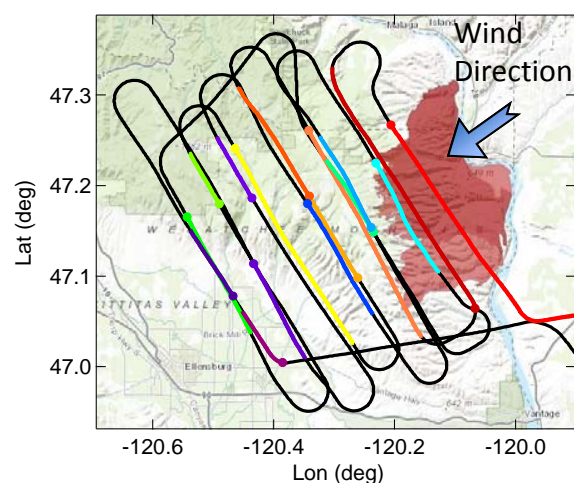
- Organic particles distinguished by shape, composition, high viscosity, lack of crystallinity, and possibly refractory character
- Uniquely recognized through electron microscopy; overlooked by other measurements (?)
- Size Range: 150 – 400 nm
- Can contain up to 10 mol % of non-carbon elements, mainly H, O, S
- Are one of the major identified components of brown carbon (BrC)
(e.g., Andreae & Gelencser 2006)

Evolution of Tar Balls

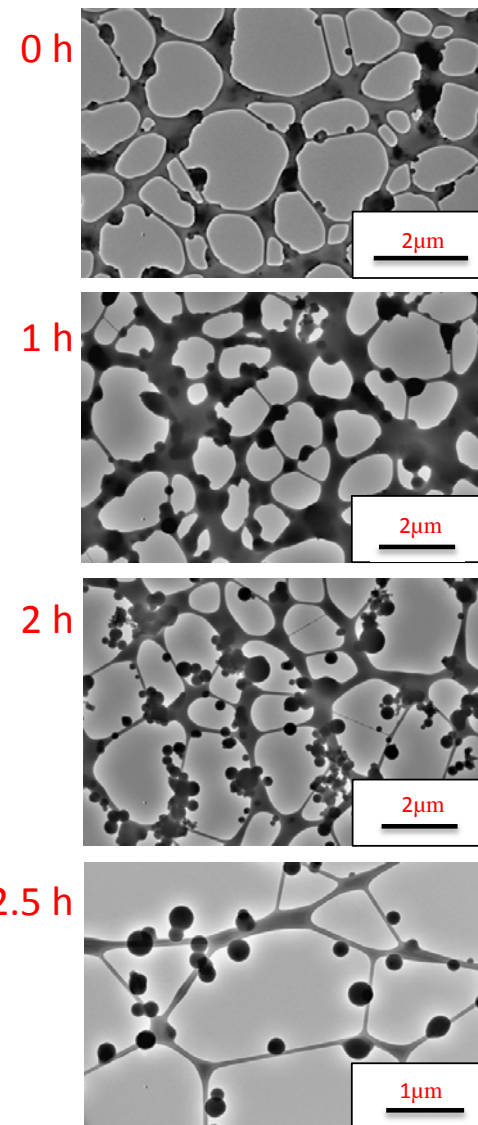
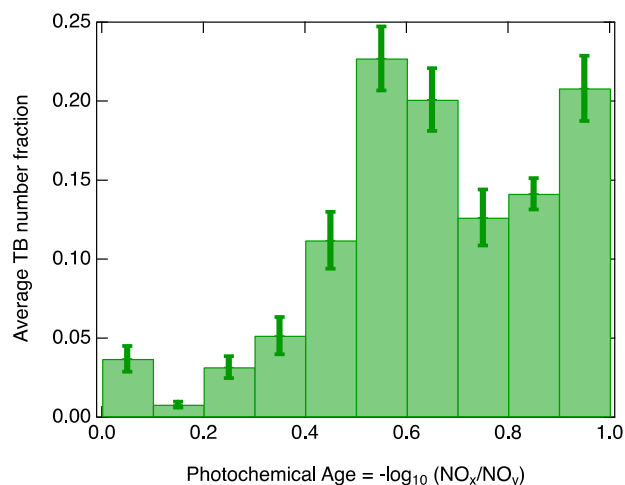
Lingering uncertainty about TB Formation mechanism

- Primary: rapid heating of primary plant emissions
- Secondary: oxidation/photolysis of POA

BBOP data shows increase in TBs number as a function of plume age



Analysis of all BBOP wildfire TEM samples supports a secondary formation mechanism



TB Fraction in a Wildfire Plume

Conclusive detection of TBs is still limited to electron microscopy (e.g., TEM)

Several TEM studies have reported very high TB fractions (>50%)

However:

Loss of volatile material can occur during storage and electron beam interrogation

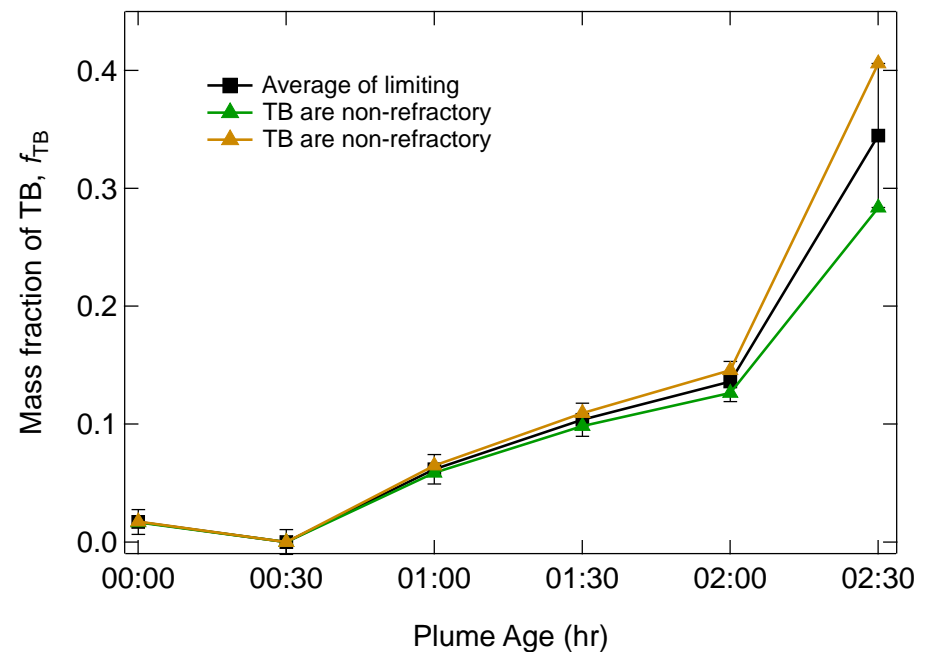
TEM-derived TB fractions overestimate the true contribution of TBs

Combine TEM, AMS, and SP2 to provide estimate the TB mass fraction in the plume

- TEM: M_{TB}/M_{soot}
 - AMS: M_{org} , M_{inorg}
 - SP2: M_{rBC}
- rBC (SP2) = soot (TEM)

TBs may be refractory and not detected by AMS

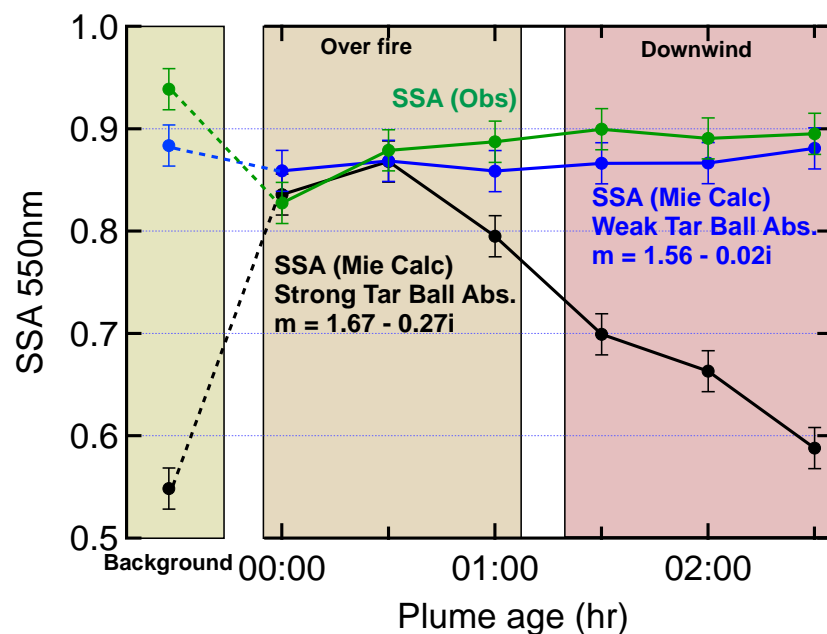
First report of TB mass fraction, f_{TB} , in plume



Optical Properties of Tar Balls

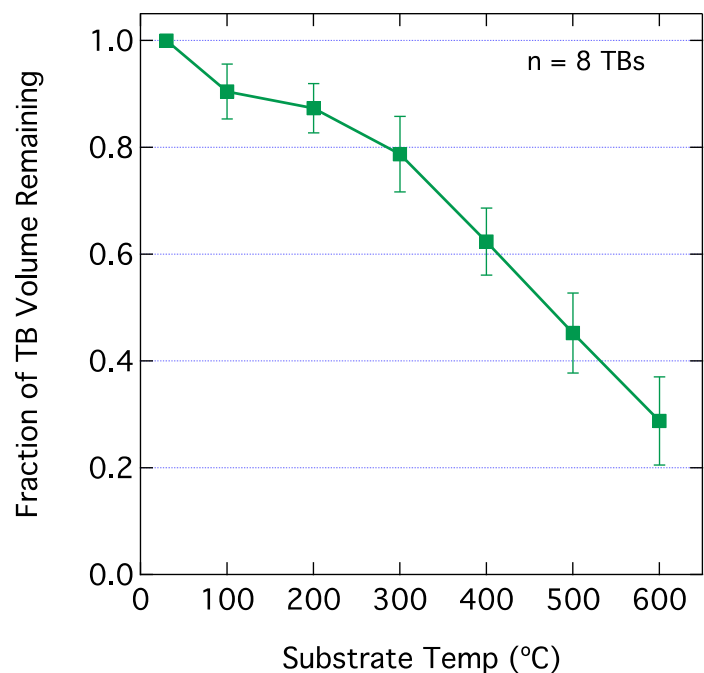
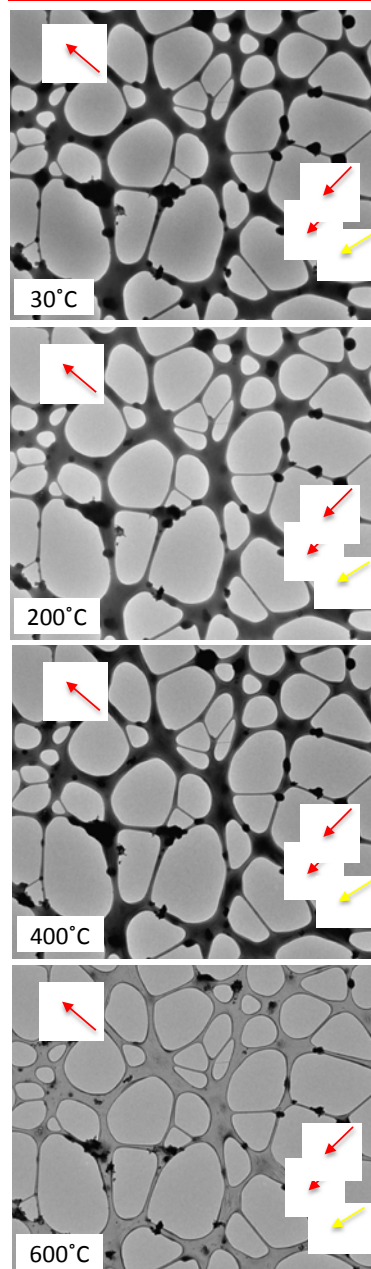
100x difference in
imaginary component

Refractive Index	TB Source/Analysis Technique	Wavelength (nm)	Reference
1.67 - 0.27i	ACE-Asia Field campaign - Electron Energy Loss Spectroscopy	550	Alexander et al., 2008
1.84 - 0.12i	Tar-water emulsion - Light Absorption	550	Hoffer et al., 2015
1.56 - 0.02i	YACS field campaign - OC/EC ratio & Scattering	632	Hand et al., 2005
1.80 - 0.007i	Ponderosa Pine - Light Absorption	532	Chakrabarty et al., 2010
1.75 - 0.002i	Alaskan Duff - Light Absorption	532	Chakrabarty et al., 2010

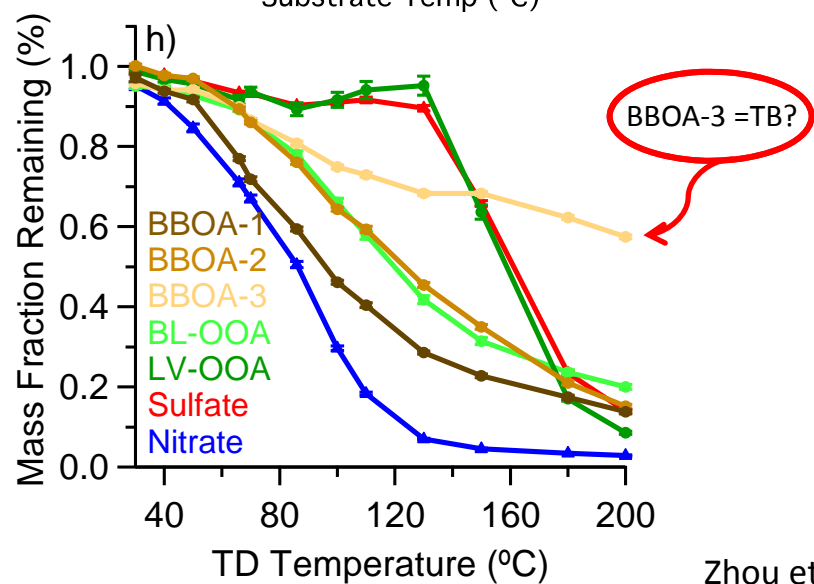


An SSA derived from optical measurements is consistent with Mie calculations that assume that TBs are weak absorbers (1.56-0.02i).

Refractory Properties of Tar Balls



Thermal experiments suggest that TBs may not be detected by AMS



Zhou et al., 2017

Research Questions

- Secondary production of Tar balls.
 - What is the role of photochemistry in the production of Tar balls?
 - Is there a pronounced diurnal cycle in the production of TBs?
- Tar ball mass fraction in smoke plumes
 - How variable is the Tar Ball mass fraction in wildland fires.
- No TBs observed in Ag-burns, but observed in most wildland fires studied during BBOP.
 - Is there a fuel source dependence on the production of Tar balls?
- Laboratory experiments to augment field observations.
 - How representative of laboratory-generated TBs to those observed in the field?
 - What is the origin of the imaginary component of the TB refractive index?
 - What is the refractory character of TBs? Impact on detection by other techniques.

