Laboratory Studies of Black Carbon Particles: Characterization and Atmospheric Processing

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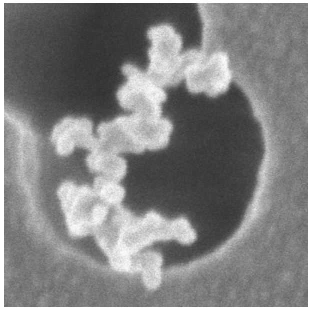
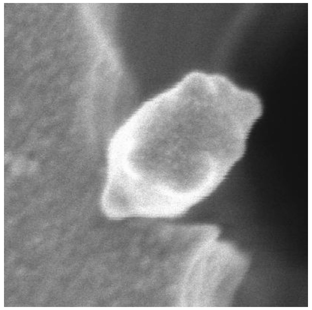
Soot Particles

- Products of incomplete combustion. Large anthropogenic contribution to atmospheric loadings (e.g. internal combustion engines, fossil fuel, biomass, biofuel, etc.)

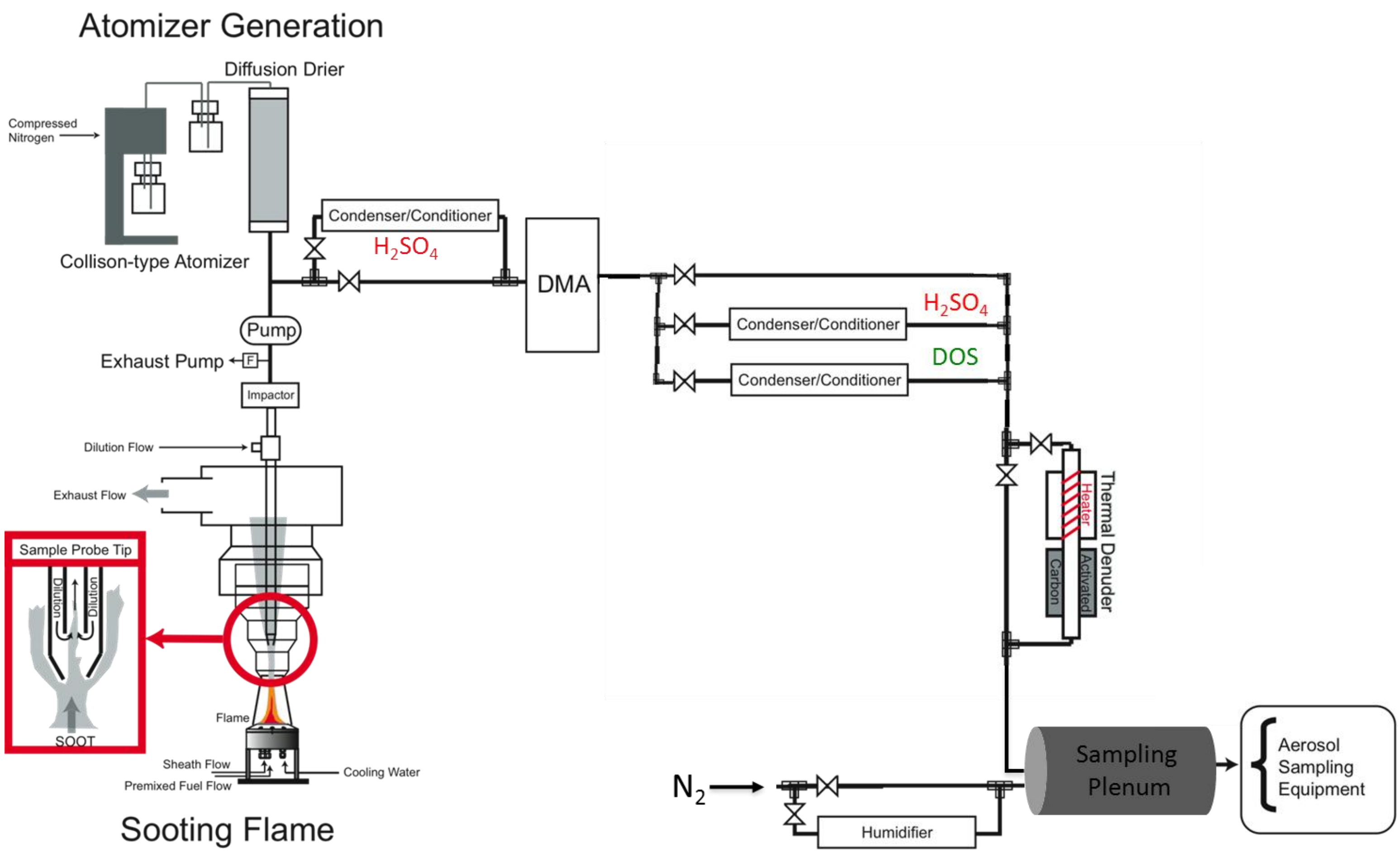
- Soot particles are complex, nonspherical, and composed of heterogeneous mixtures (e.g. ***refractory carbon***, polycyclic aromatic hydrocarbons, incomplete combustion products, engine oils, photochemical oxidation products, etc.)



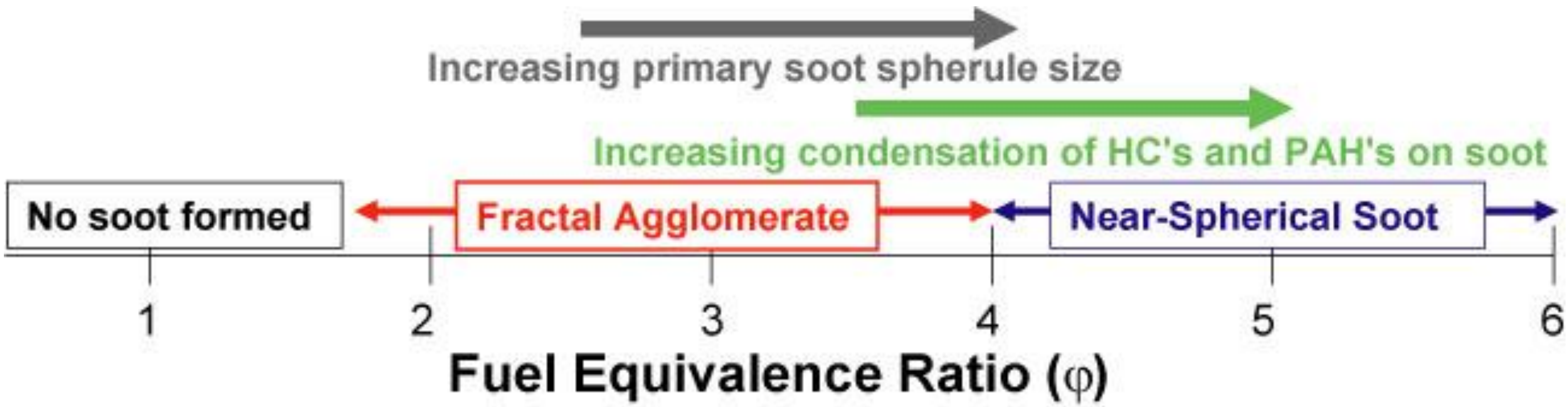


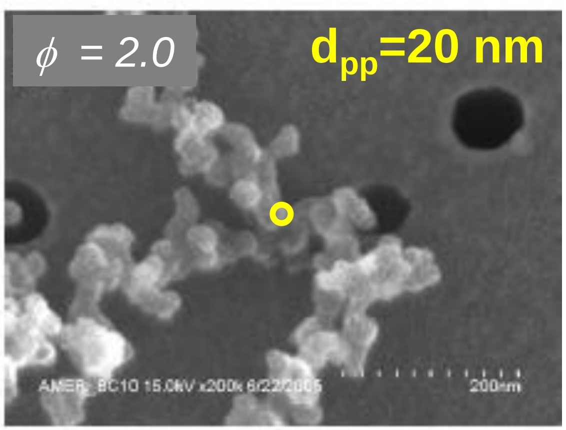
- Soot particles absorb across the solar/terrestrial spectra with implications for atmospheric radiation balance, snow albedo, and clouds

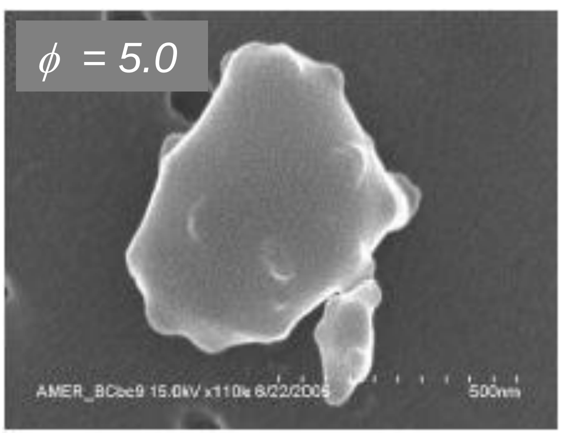
Laboratory setup for carbonaceous particle generation and characterization



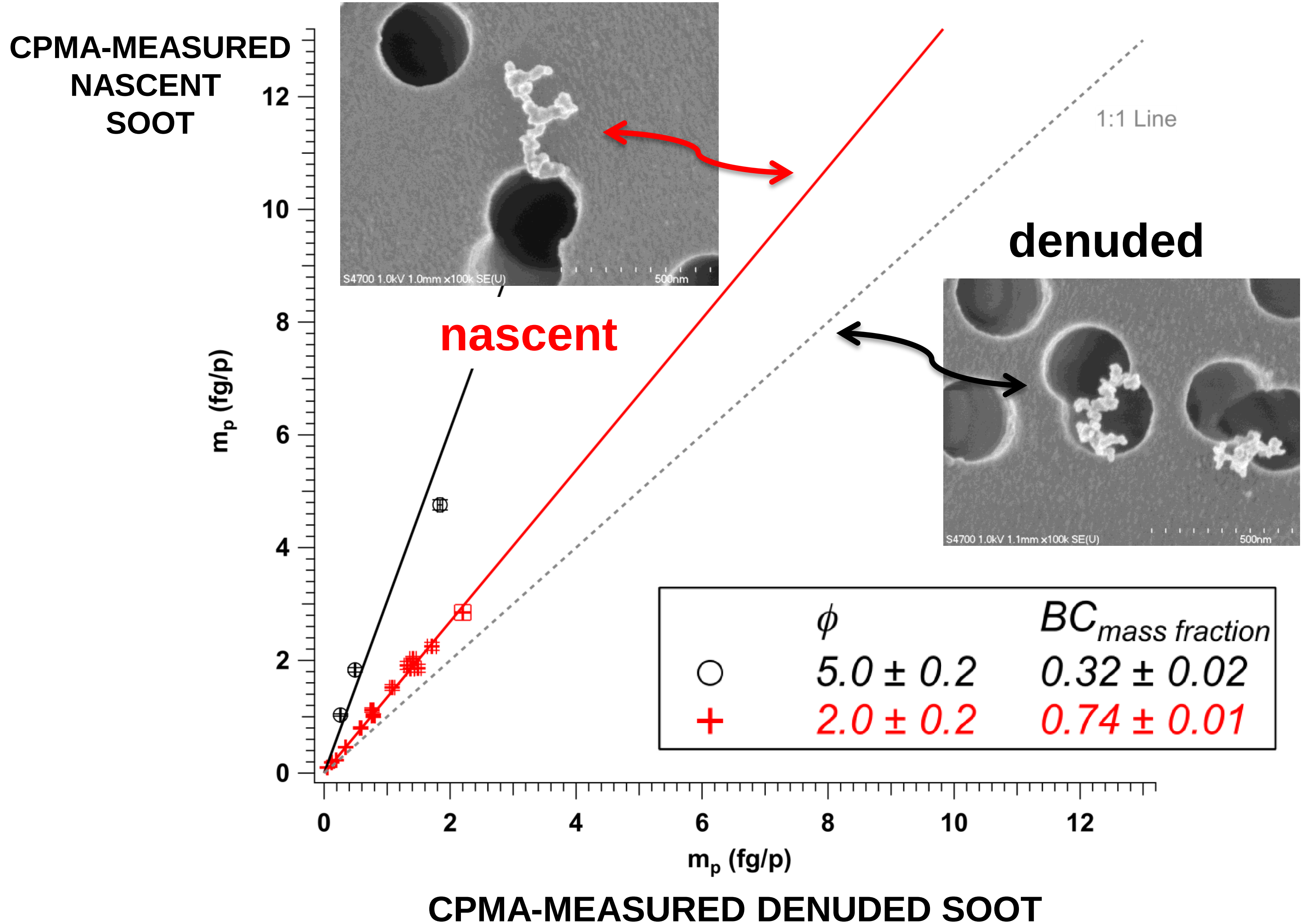
Monodisperse Soot Generation





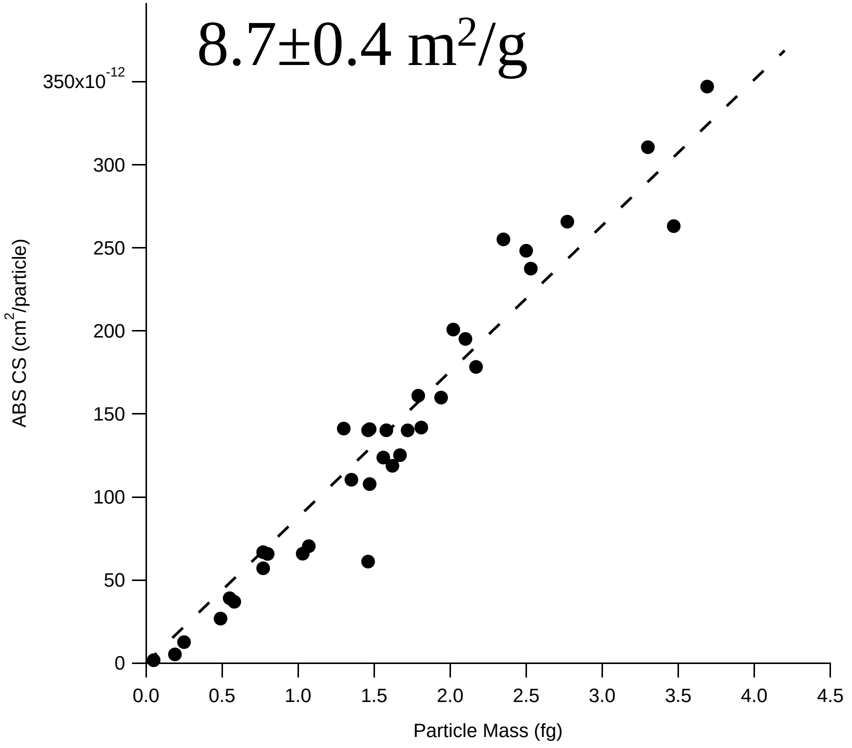


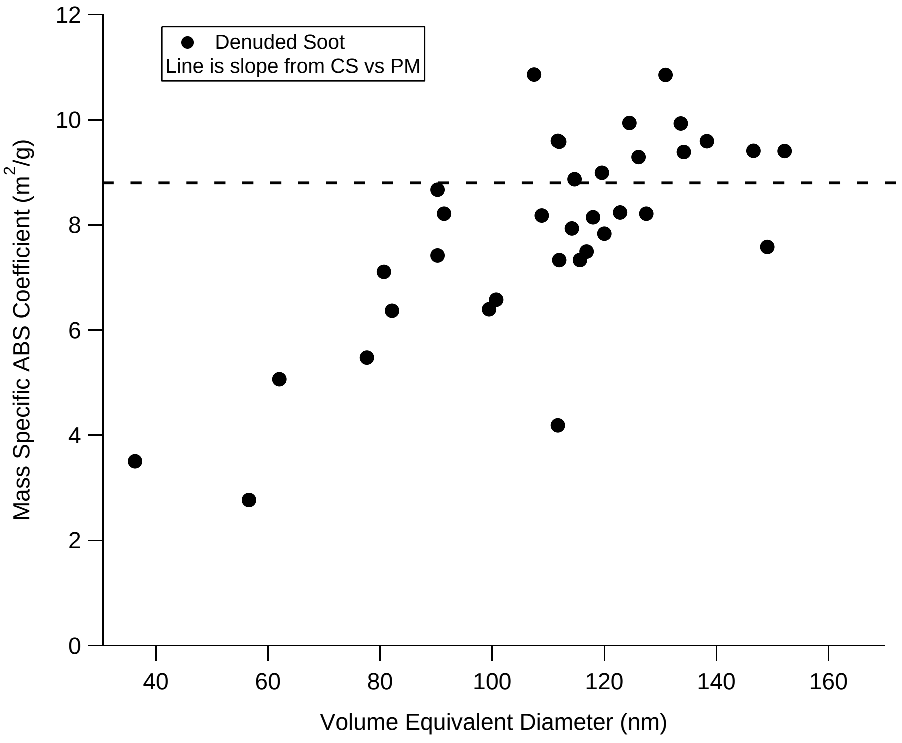
‘Efficient’ Combustion Soot



Mass Specific Absorption Efficiency

λ = 532 nm





- Mass specific absorption efficiency = 8.7±0.4 m^2/g
- Size dependence

Lack, Cappa, et al.

Mass Specific Absorption Efficiencies

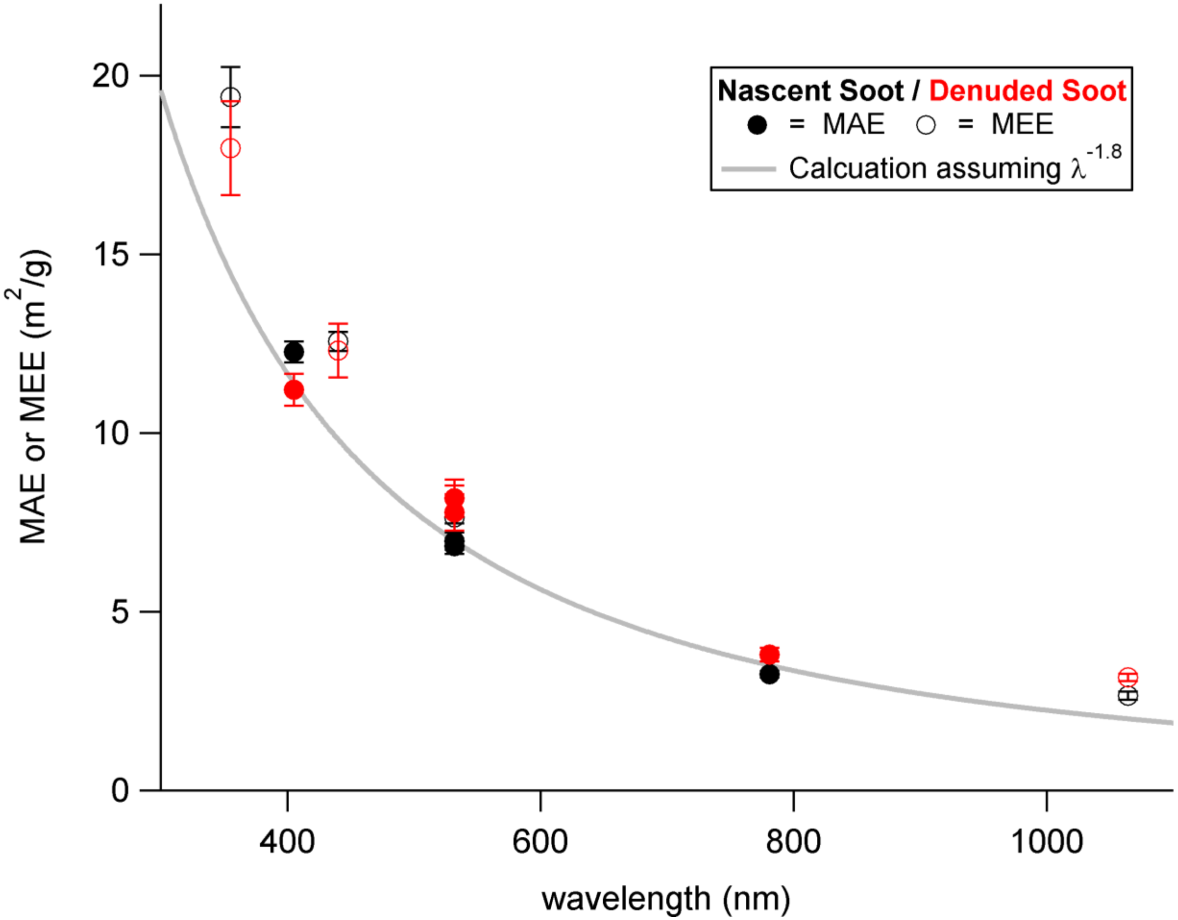
Soot Type	MAE (m^2/g)	Wavelength (λ nm)
Denuded	8.7 ± 0.4	532
Nascent	7.5 ± 0.5	532

$$MAE \; \alpha \; m_p^{-1}$$

- Denuded/Nascent m_p ratio = 0.74
- Nascent/Denuded MAE ratio = 0.86

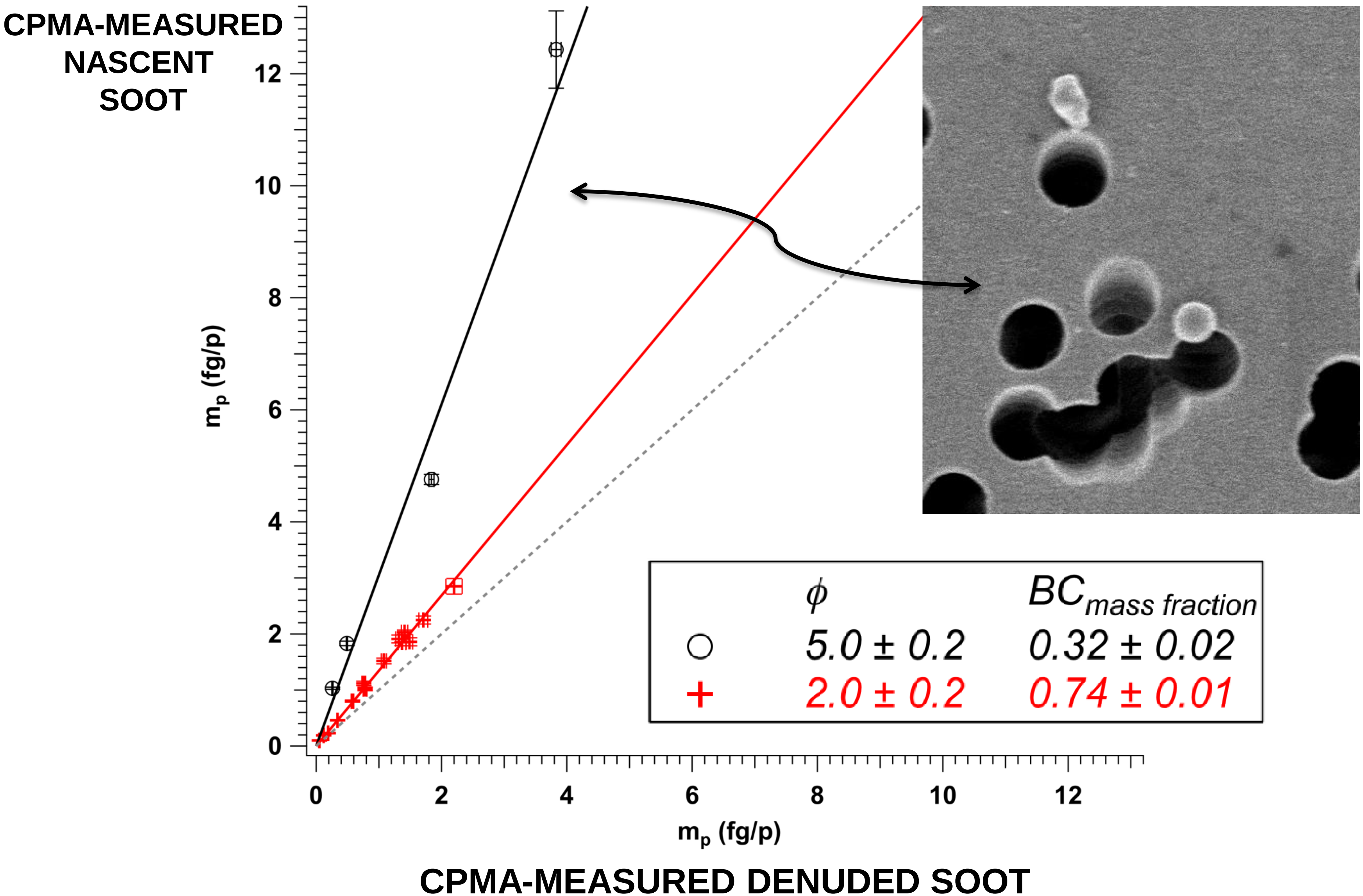
- The measured variation in nascent/denuded MAE is approximately the same as the variation in the per particle mass (i.e. mass lost during denuding)

Wavelength dependence of flame soot



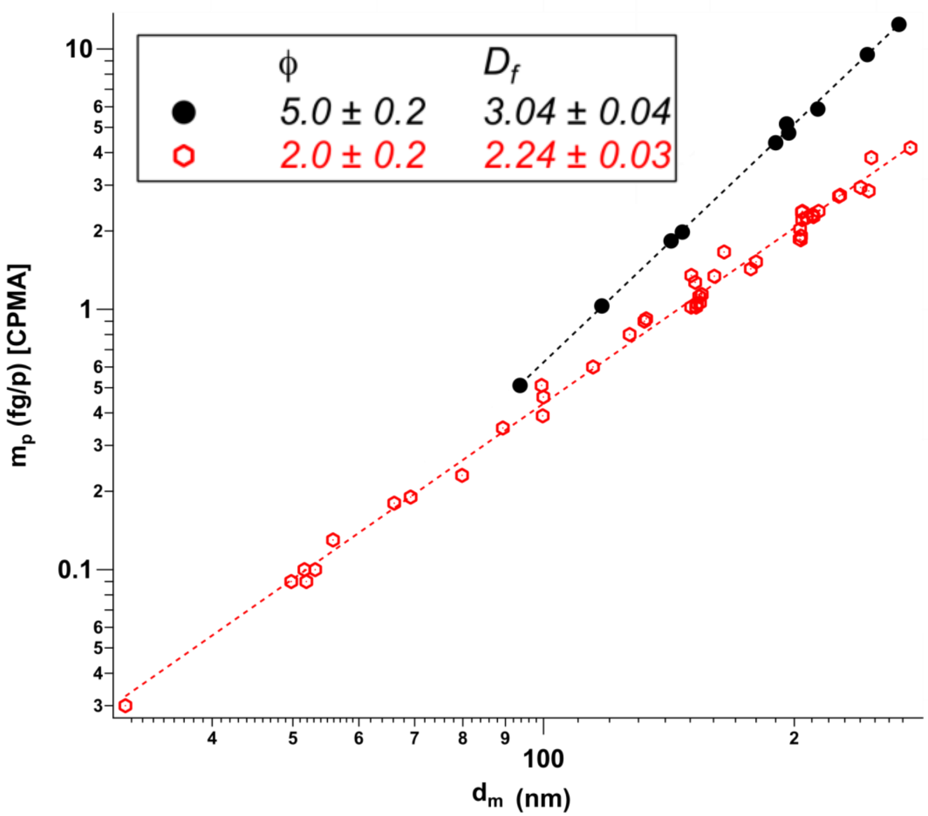
- Mass specific absorption efficiencies exhibit increases greater than $1/\lambda$ with decreasing wavelength

'Inefficient' Combustion Soot

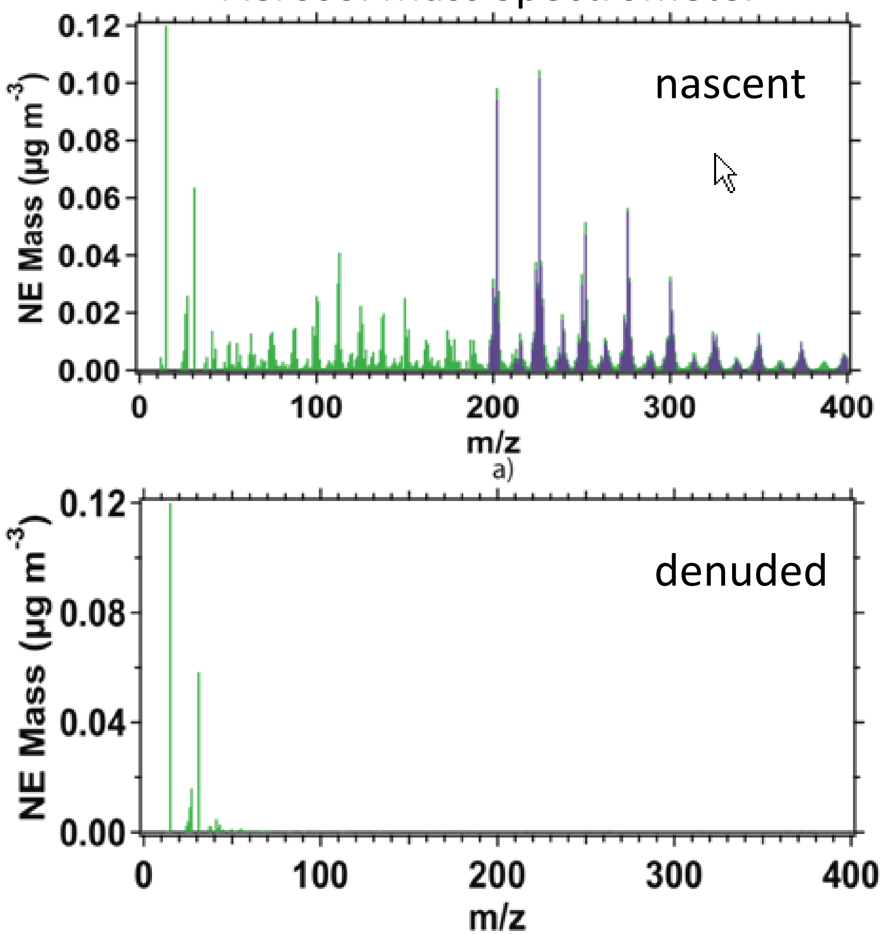


Morphology and Chemistry

$$m_p = C \cdot d_m^{Df}$$

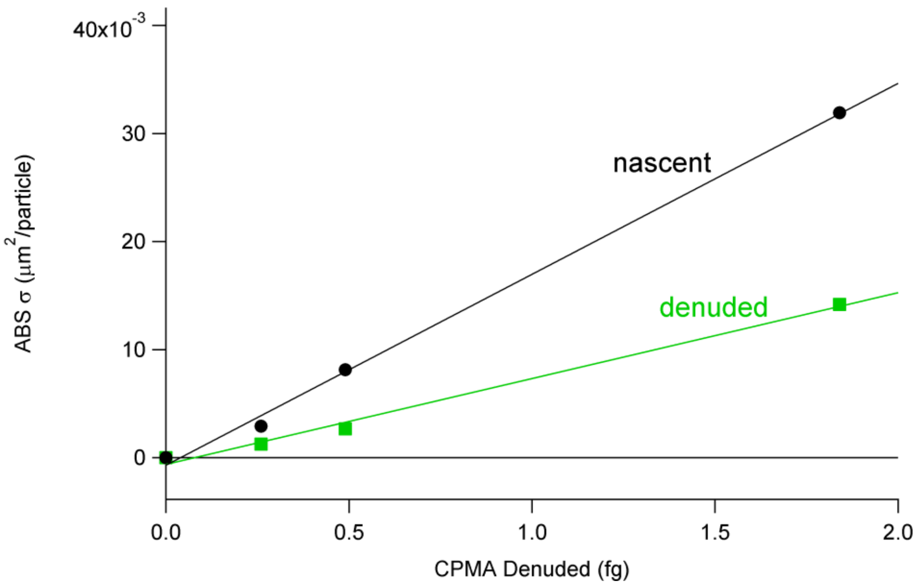


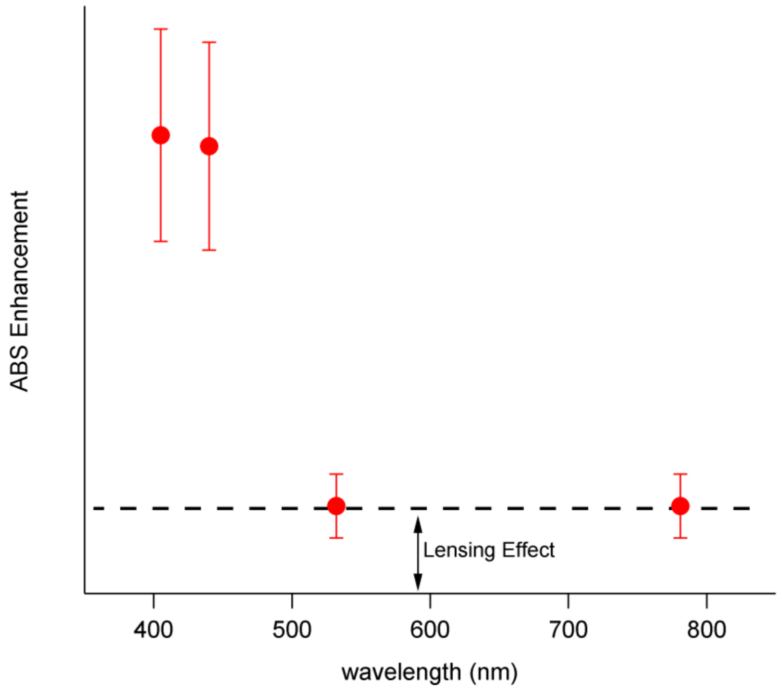
Aerosol Mass Spectrometer



- Soot particles coated to spherical particles
- Coating material is mainly conjugated Polycyclic Aromatic Hydrocarbons (PAH)

ABS Enhancement and Evidence for Absorbing Organics





- Absorption increases due to coating
- Long wavelength results likely show lensing effects
- Wavelength dependence strongly suggests absorbing organics (i.e. brown carbon)

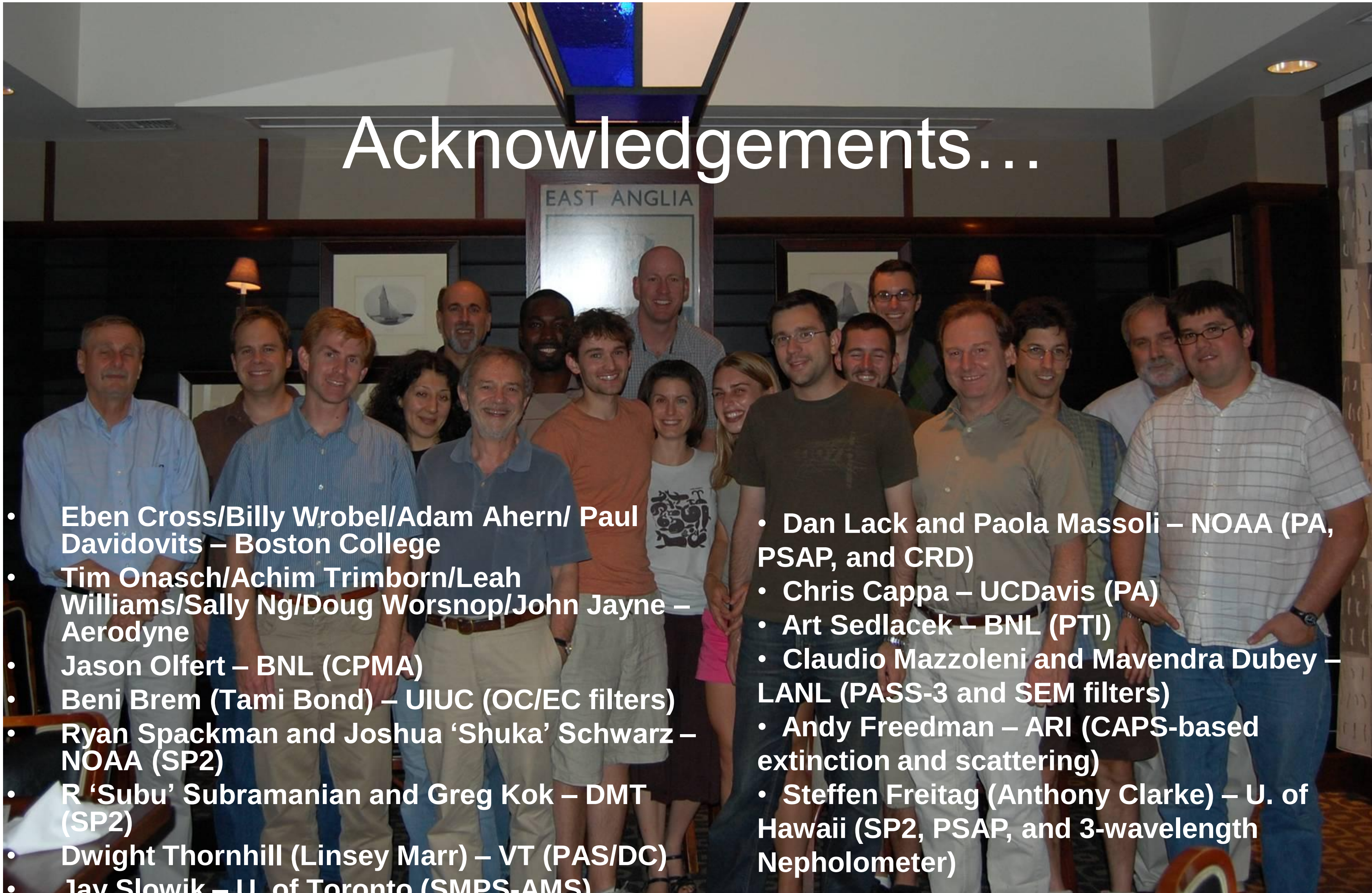
Mass Specific Absorption Efficiencies

Soot	MAE (m^2/g)	MAE Calculated	Wavelength (l nm)	Reference
Refractory Carbon	8.7 ± 0.4	-	532	This work
'Efficient' combustion	7.5 ± 0.5	6.5	532	This work
'Inefficient' combustion	6.5 ± 1	2.8	532	This work
Variable	8.0 ± 1.2		550 converted to 532	Bond and Bergstrom, 2006

- Calculated MAE accounts for particle mass increase due to nonrefractory coating only
- Difference between the measured and calculated MAEs due to lensing at 532nm and longer wavelengths

Summary

- Refractory carbon MAE = 8.7 m^2/g at 532 nm from ethylene flames
- Optical properties at 532 nm of ‘efficient’ and ‘inefficient’ nascent soot exhibit similar, though lower, MAE’s due to off-setting effects:
  - Increasing nonabsorbing particulate mass
  - ABS enhancements due to lensing
- Wavelength dependence of refractory carbon is described by an inverse power relation with power ~2
- ‘Inefficient’ combustion generates absorbing (400-450 nm) organic compounds (e.g. PAH’s) that complicates a simple wavelength dependence



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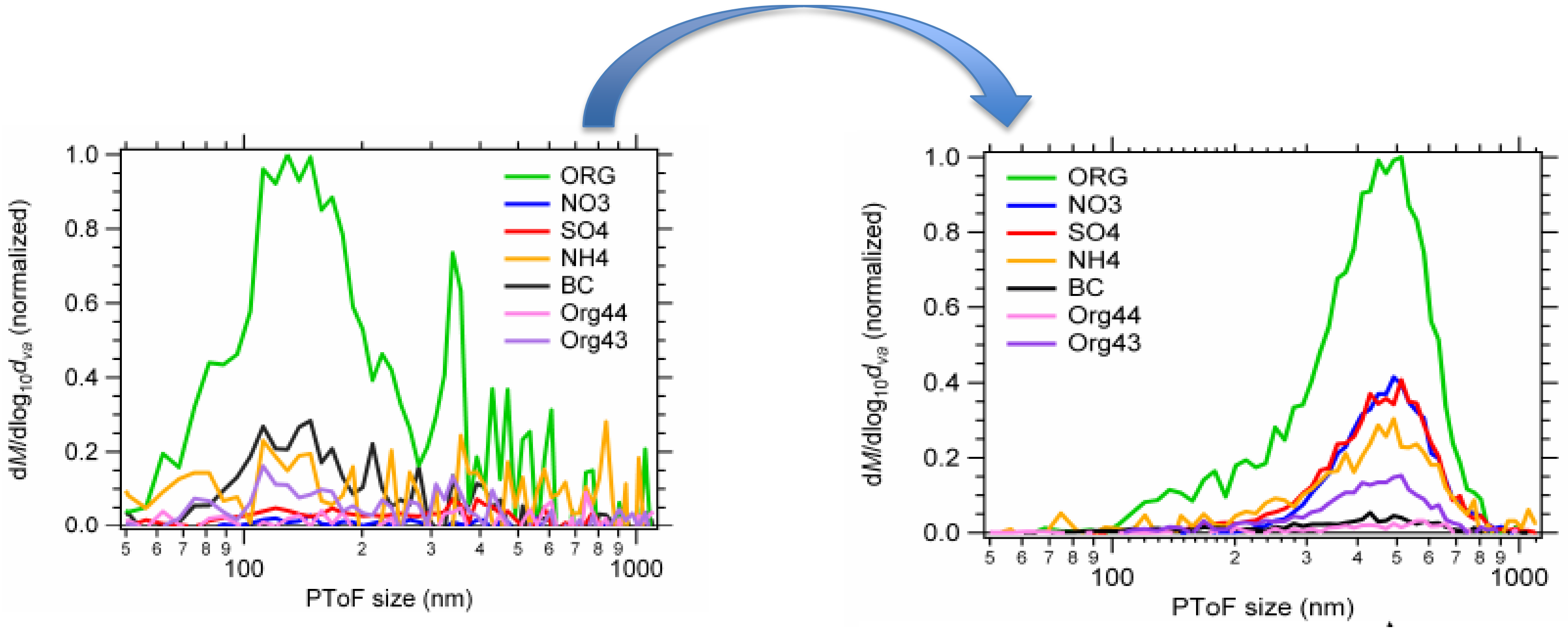
DOE ASR Funding

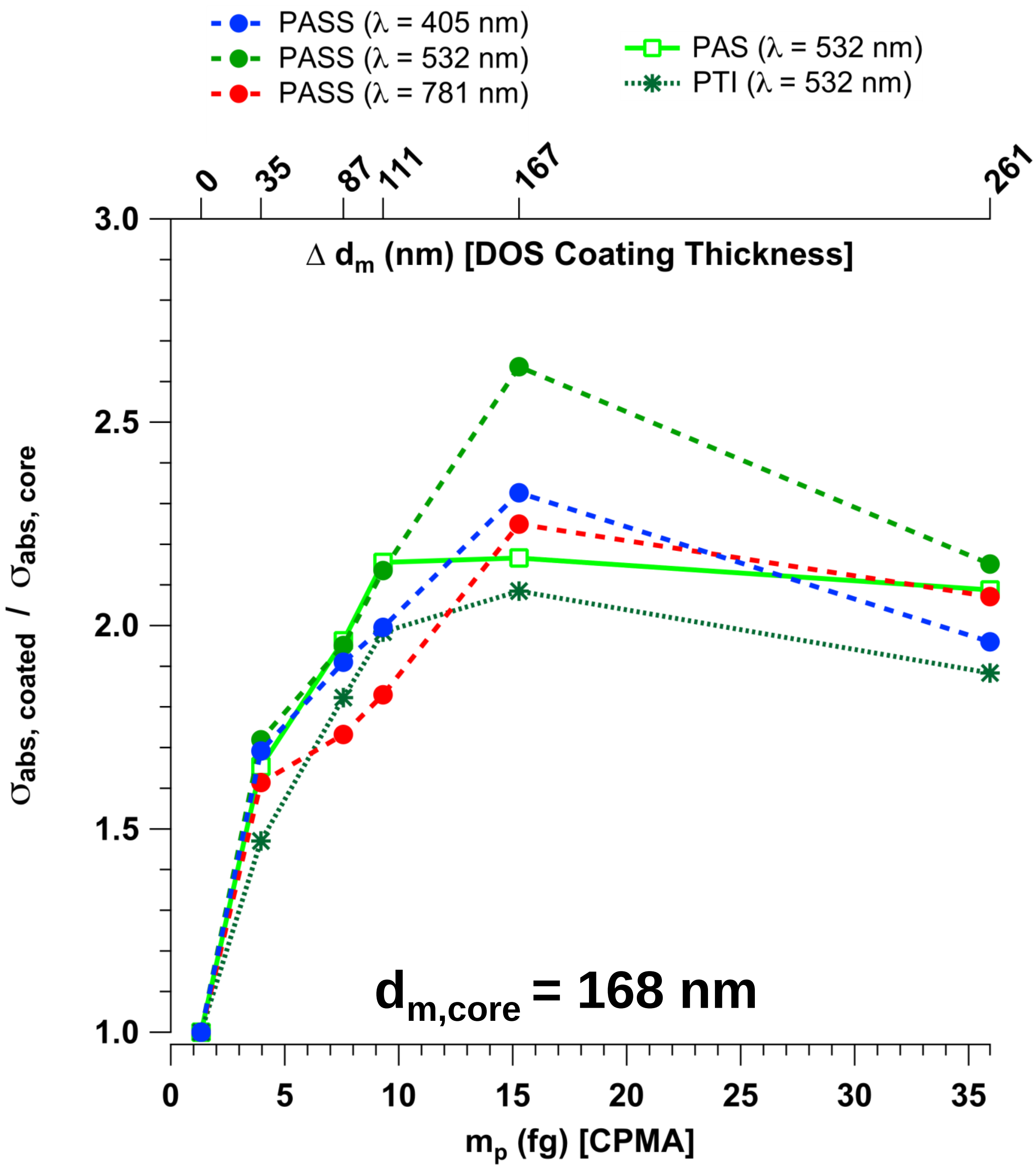
Effects of Atmospheric Aging



- New instruments (SP-AMS) starting to provide valuable information on atmospheric aging, soot coating levels and chemistry

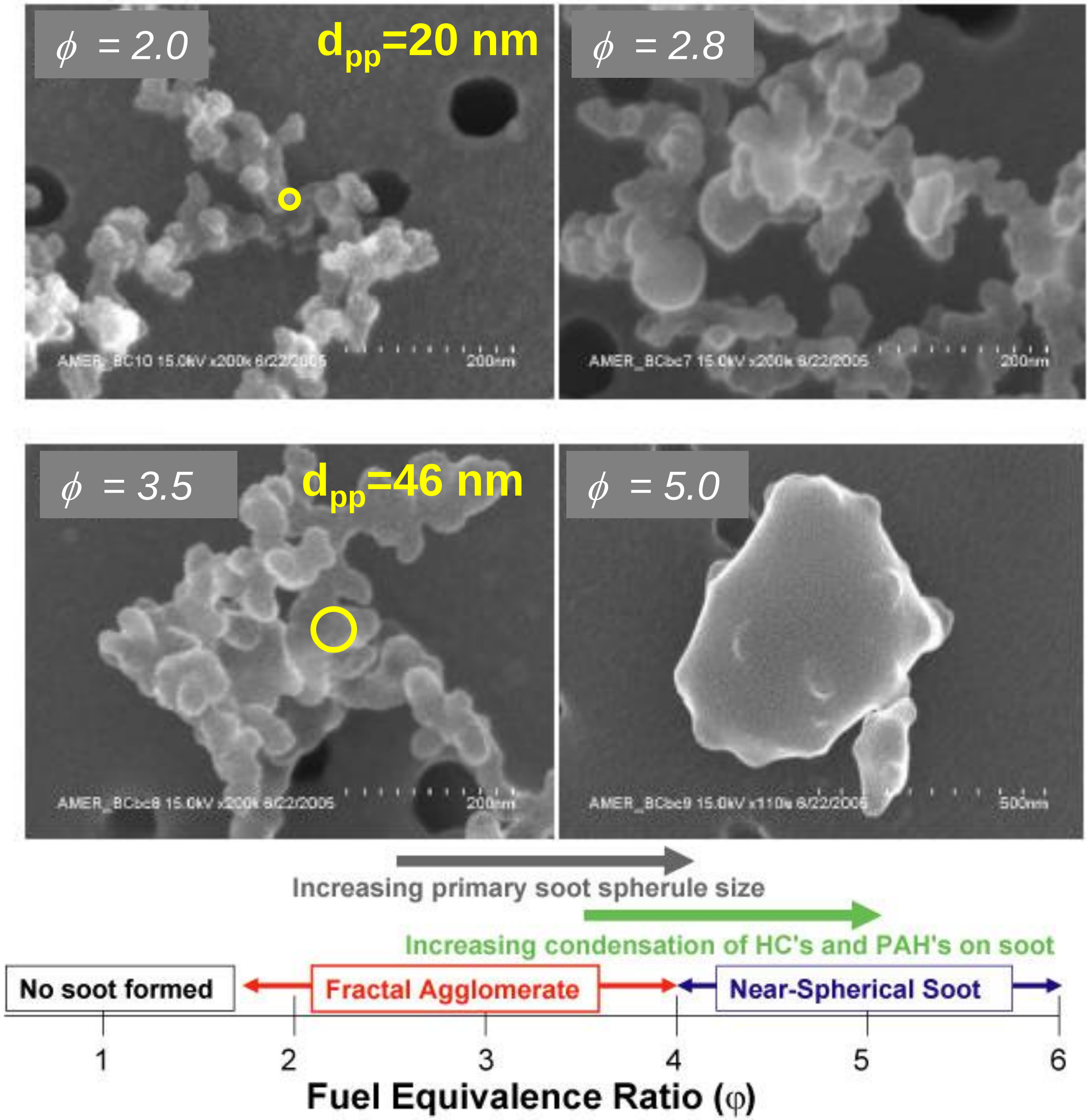
Extra Slides

Absorption Enhancement



$\phi = 2.0 +/- 0.2$

Monodisperse Soot Generation



Operating Parameters

$2.0 < \varphi < 5.0$

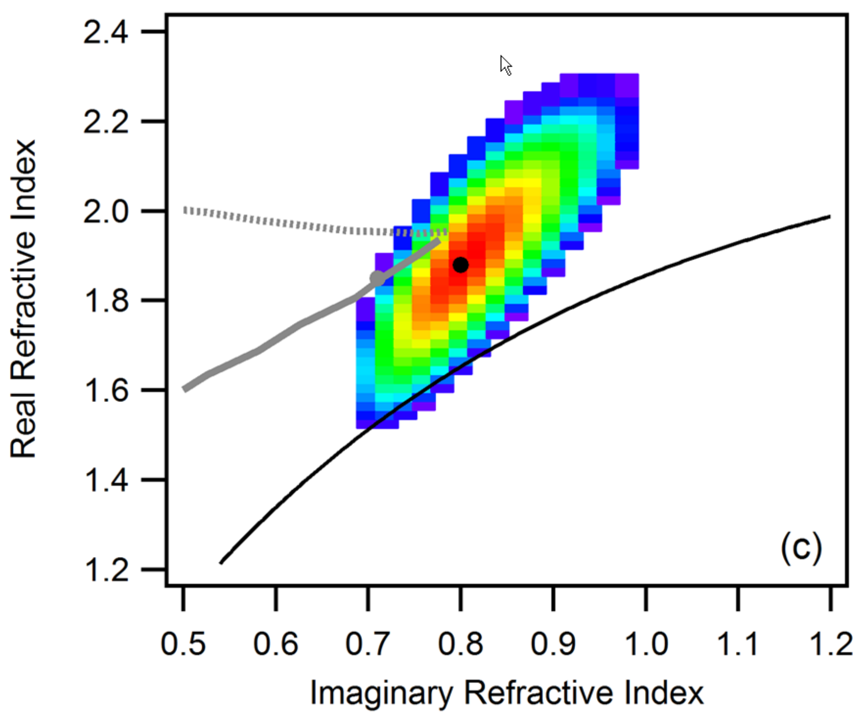
$30 < D_{mobility}$(nm) < 500

$15 < D_{pp}$ (nm) < 55

0 < Coatings (nm) < 250

Complex Refractive Index

λ = 532 nm

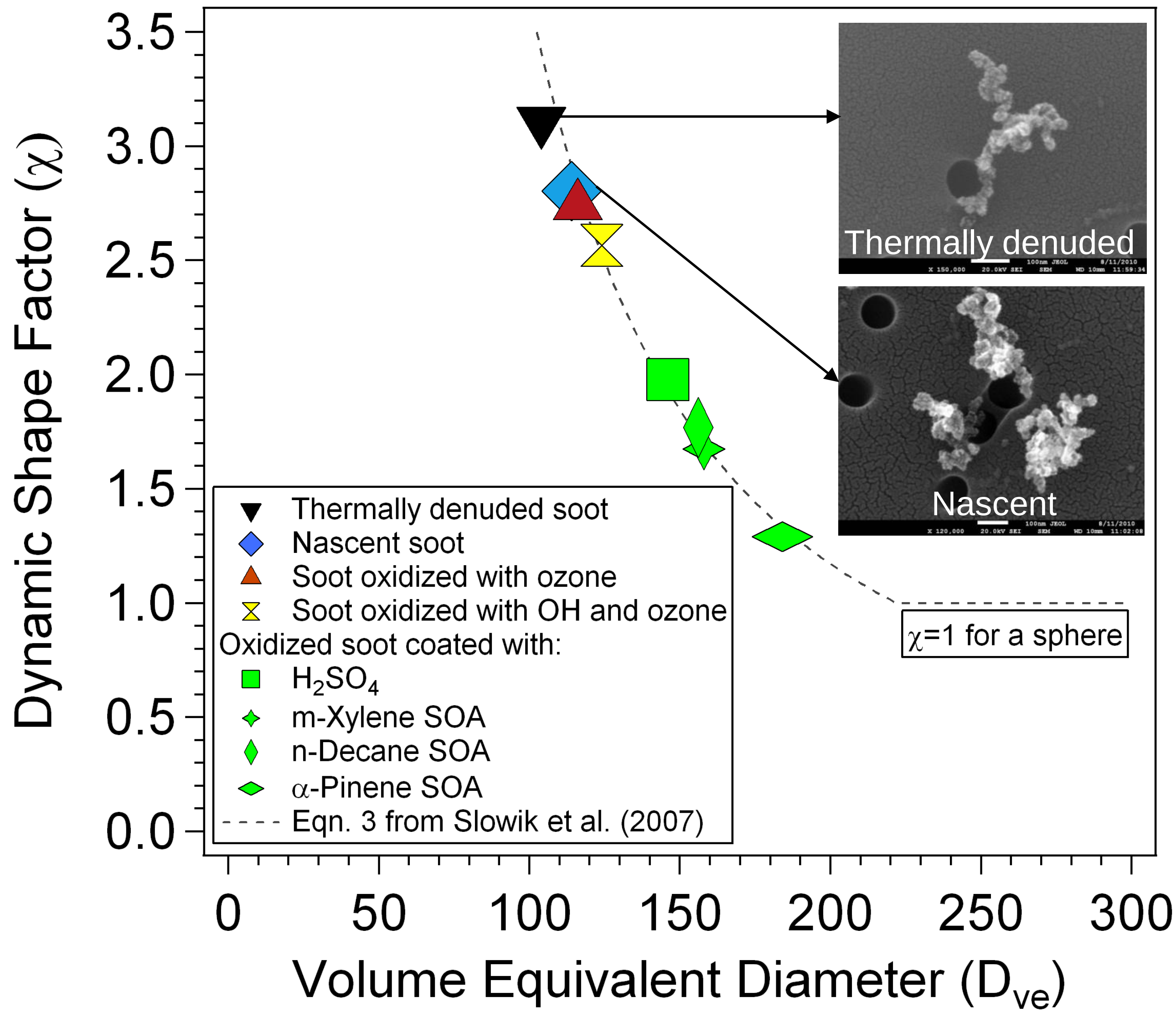


Soot Type	*n*	Reference
Nascent	1.88 + 0.80*i*	This work
Variable	1.95 + 0.79*i*	Bond and Bergstrom, 2006

- Mie Theory, fitting measured MAE and MEE
- Nascent soot

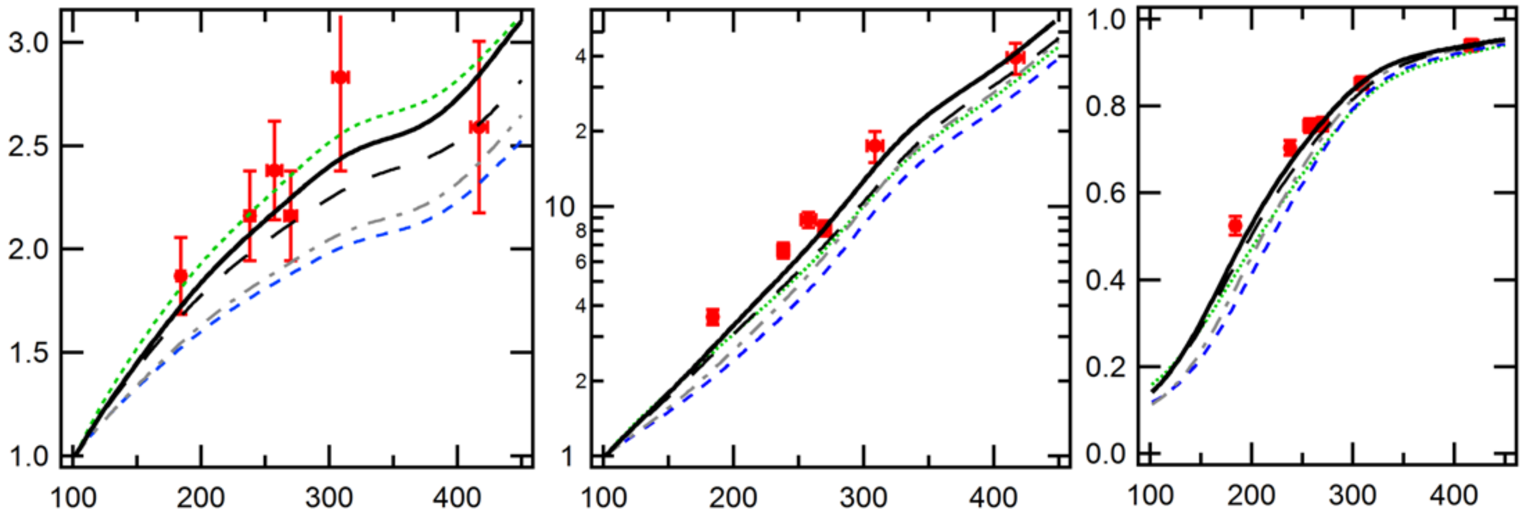
Cappa, Lack, et al.

Effects of Coating on Soot Morphology



Coating Soot Particles

Absorption Enhancement | Extinction Enhancement | Single Scatter Albedo



Diameter (nm) | Diameter (nm) | Diameter (nm)

- Nascent Soot core (Volume Equivalent Diameter = 102 nm)
- Derived Complex Refractive Index
- Di-octyl sebacate coating
- Mie Core-Shell Theory appears to model measurements

Cappa, Lack, et al.