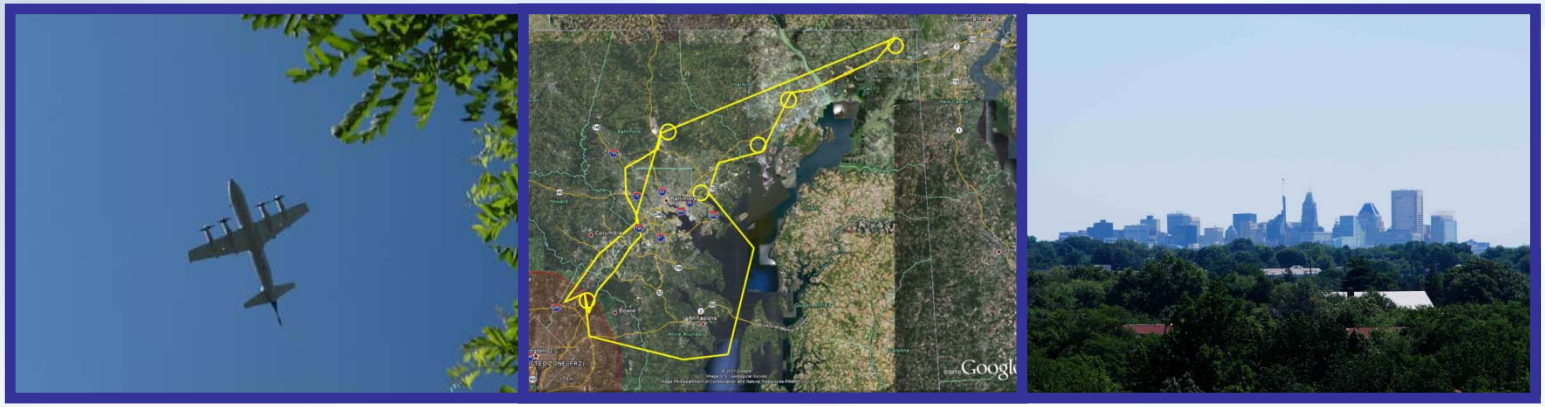
Aerosol Composition and Variability in Baltimore Measured during DISCOVER-AQ



Andreas Beyersdorf, Luke Ziemba, Suzanne Crumeyrolle, Lee Thornhill, Gao Chen, Edward Winstead, Glenn Diskin & Bruce Anderson

• NASA Langley Research Center •

Robert Chatfield

• NASA Ames Research Center •

Vijay Natraj

• Jet Propulsion Laboratory •

Motivation

Geostationary satellite instruments allow for tropospheric air quality measurements

- GEO-CAPE
- TEMPO

Satellite measurements must be related to surface conditions

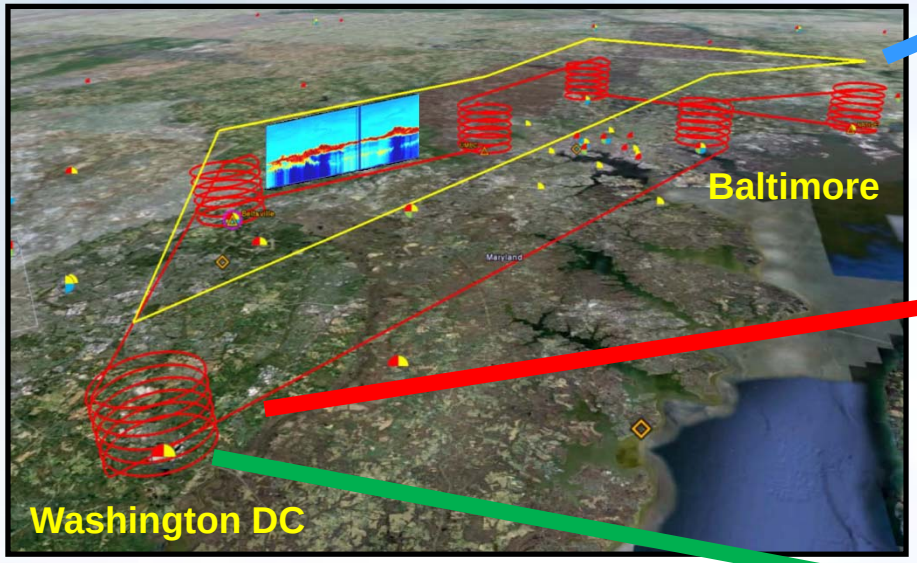
- How do column measurements relate to the near surface?
- What are the affects of meteorological conditions on retrievals?
- What spatial variability is there?

DISCOVER-AQ = Deriving Information on Surface Conditions from COlumn and VERtically Resolved Observations Relevant to Air Quality

- Multi-year, multi-location campaign
- Particulate and gas-phase measurements
    - High-altitude satellite instrument simulators
    - Aircraft profiling
    - Ground sites

Sampling Strategy

Baltimore, MD and Washington D.C. in July 2011



14 flight days in 29 days

- **UC-12** performed 3-4 circuits per flight over the region at high altitude
- **P3B** performed three circuits per flight
  - profiles over six instrumented ground sites
    - Total of 253 profiles and 41 circuits
  - Additional low level legs over freeways and the Chesapeake Bay

UC-12

- HSRL; vertical distribution of aerosol
- ACAM

P3-B

- LARGE; in situ aerosol optical and microphysical measurements

Ground Sites

- 6 supersites; spiral locations
- 7 additional sites

L. Brent Poster
(GC21B-0968)

Sampling Strategy

Remote Measurements

- Primary retrieval is aerosol extinction
- Decreased sensitivity near clouds and the ground
- Measurements are at **ambient RH**

In Situ Airborne Measurements

- Can measure a wider range of properties (size, composition)
- Inlet effects – aerosol loss & **reduced RH**
- Can modify the RH and correct to ambient conditions
- Cannot sample the lowest 500 ft

Ground Measurements

- Spatially limited
- Most measurements ($PM_{2.5}$) are at **dry** conditions

UC-12

- HSRL; vertical distribution of aerosol
- ACAM

P3-B

- LARGE; in situ aerosol optical and microphysical measurements

Ground Sites

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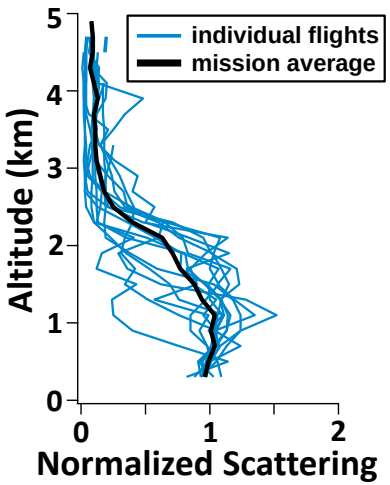
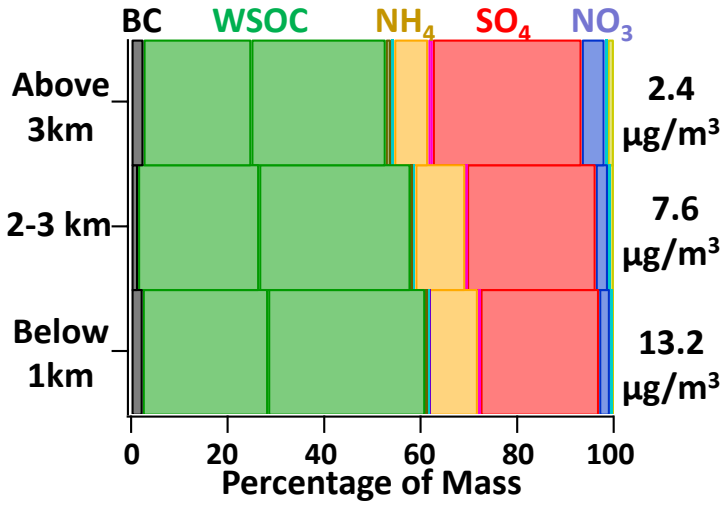
- 7 additional sites

LARGE - In Situ Aerosol Measurements

In Situ Measurements

- Aerosol concentration
- Aerosol size: 10 nm – 3µm
- Scattering (dried and at 80% RH) – corrected to ambient RH
- Absorption
- Composition - black carbon, inorganic compounds & water-soluble organics (WSOC)
  - missing mass = insoluble organics (~30% of mass)
- RH measurements made by a diode laser hygrometer and frost point hygrometer



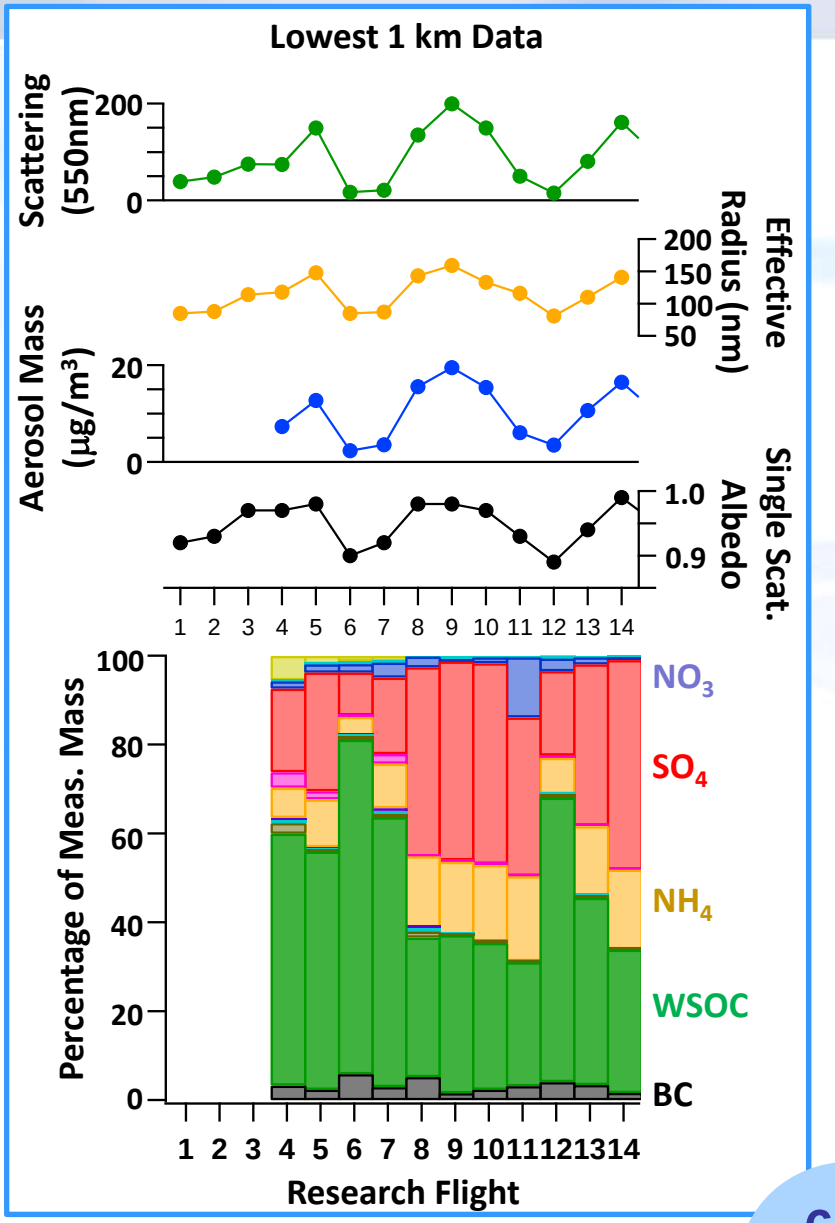


- 2 km aerosol layer
  - 36% $(NH_4)_2SO_4$
  - 58% WSOC
  - 2% Black Carbon
- Free troposphere
  - increase in relative abund. of sulfate & nitrate
  - more aged air

Variation in Aerosol Composition

Distinct difference in composition with aerosol loading. High loading days had:

- Greater abundance of ammonium sulfate relative to organics
- Larger aerosols
- Higher single scattering albedo
- Consistent with aged aerosol
- Back trajectories corresponding to transport from the Midwest



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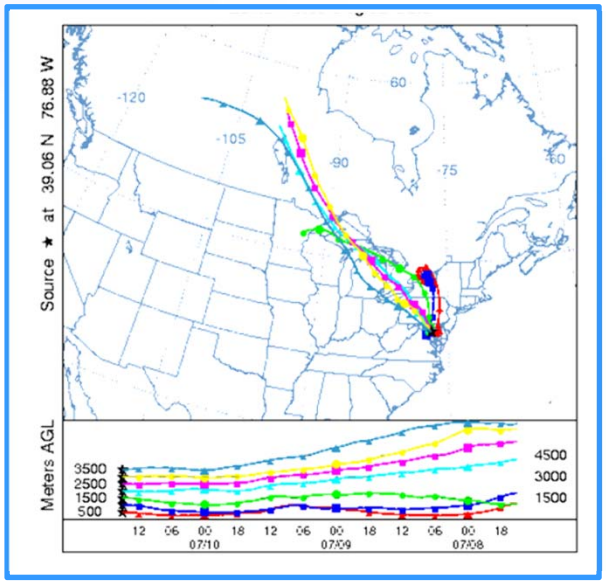
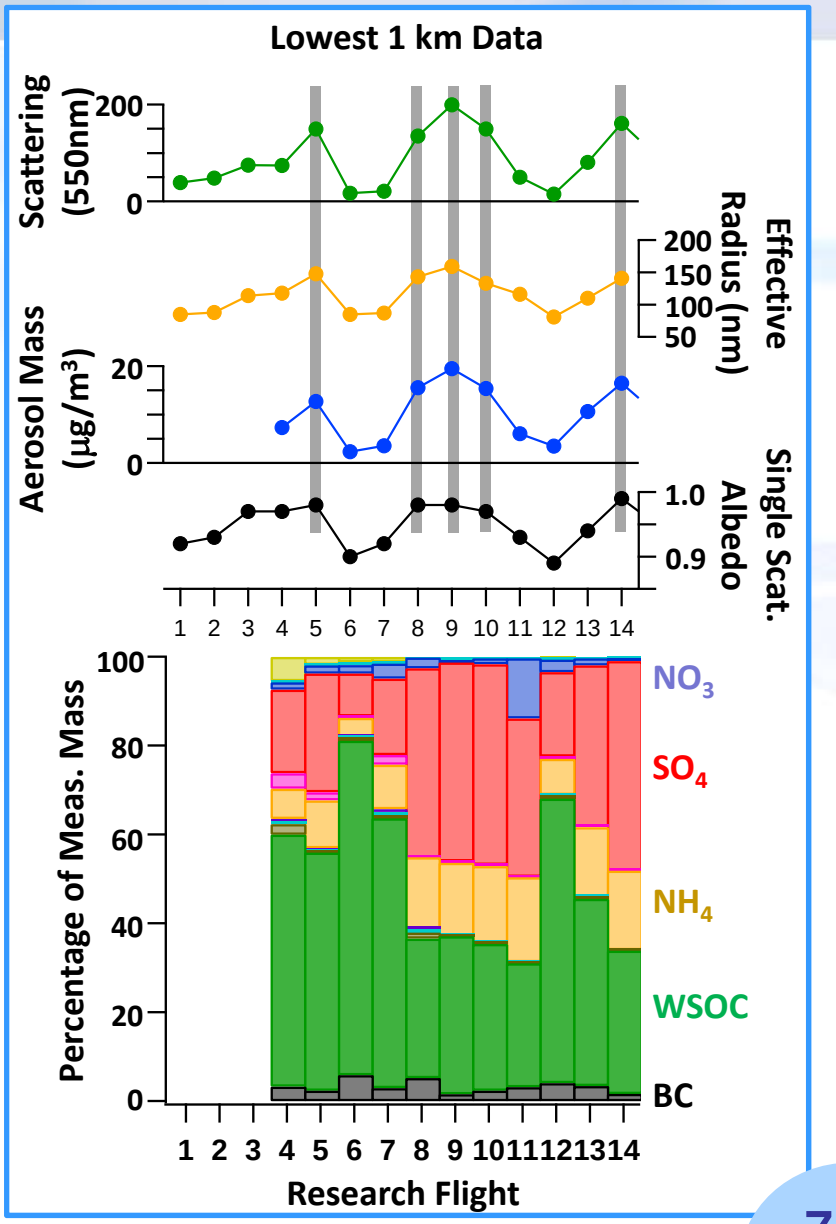




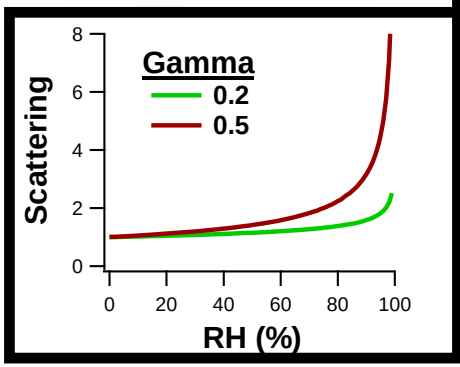
Scattering Correction

As RH increases, scattering increases due to water uptake

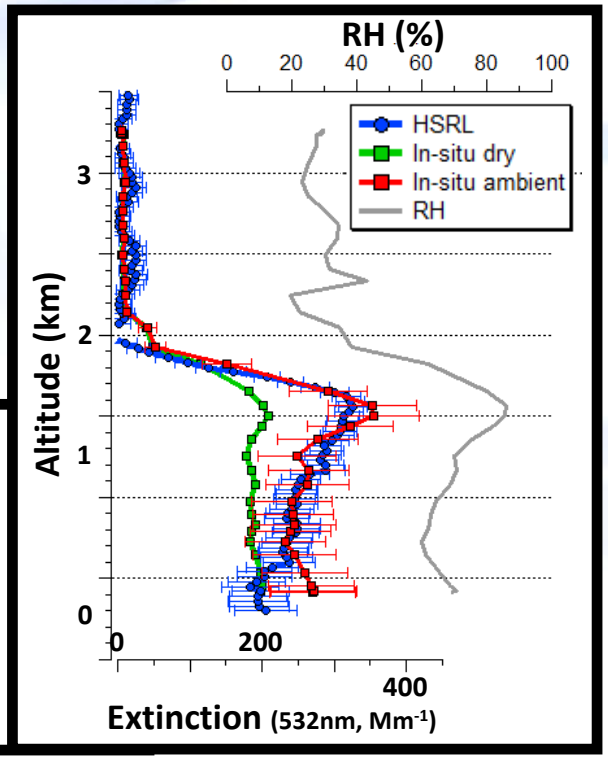
In situ aerosol scattering is corrected by measuring the scattering at two RHs

- $\sigma_{amb} = \sigma_{dry} \cdot e^{\gamma \cdot ln\left(\frac{100-RH_{dry}}{100-RH_{amb}}\right)}$
  - Gassó et al, Tellus B 2000; Quinn et al, GRL 2005; Massoli et al, JGR, 2008

- $\gamma = ln\left(\frac{\sigma_{80\%}}{\sigma_{40\%}}\right) \div ln\left(\frac{100-40}{100-80}\right)$
  - $f(RH) = \frac{\sigma_{80\%}}{\sigma_{40\%}}$



Ziemba et al., accepted, GRL

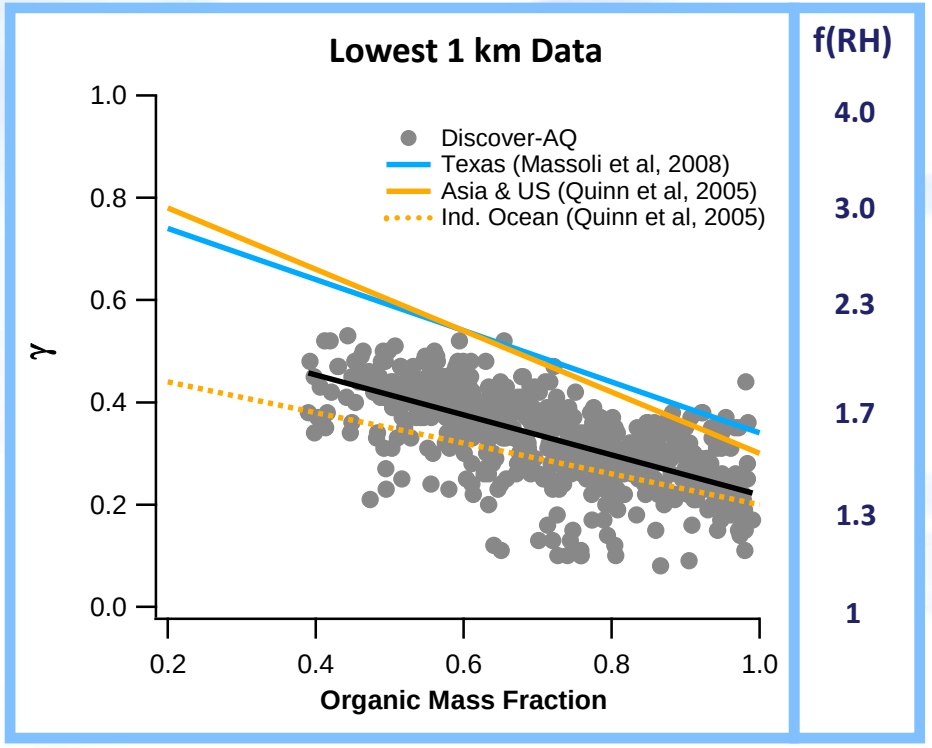


Aerosol Hygroscopicity

Gamma (γ) is dependent on composition

- $\gamma = 0.6 - 0.4 f_{POM}$
- comparable to other campaigns
  - Intermediate between fresh pollution (solid blue and yellow fits) and aged pollution (dashed fit)

No clear dependence on other aerosol properties (including size) or air mass age



Satellite retrievals are dependent on aerosol loading, composition and ambient RH

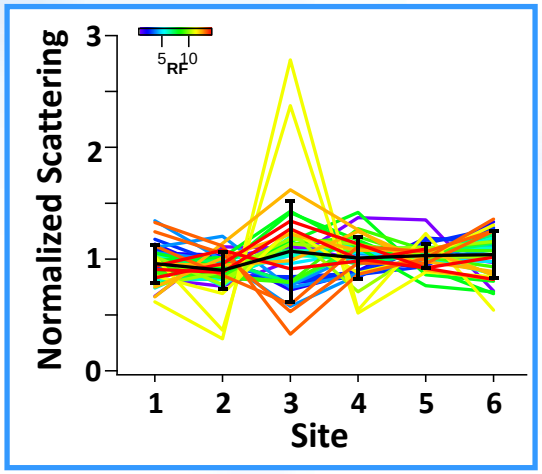
- $\sigma_{amb} = \sigma_{dry} \cdot e^{\gamma \cdot ln\left(\frac{100-RH_{dry}}{100-RH_{amb}}\right)}$

Satellite Meas. | Aerosol Loading | Composition | Ambient Conditions

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?

How much spatial variability is there in ambient aerosol scattering?

- Ambient aerosol scattering used as a proxy for AOD



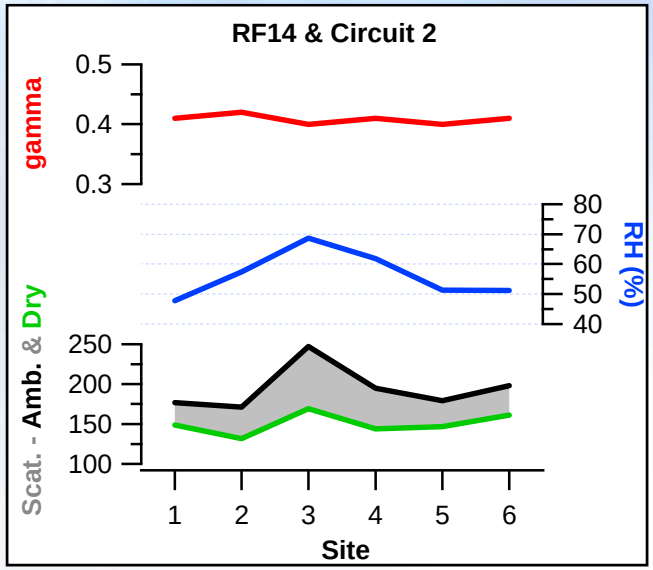
- Variability in ambient scattering for each circuit
- Average variability of 22%

What factors have the greatest effect on ambient aerosol scattering?

- $\sigma_{amb} = \sigma_{dry} \cdot e^{\gamma \cdot ln\left(\frac{100-RH_{dry}}{100-RH_{amb}}\right)}$

Satellite Meas. | Aerosol Loading | Composition | Ambient Conditions

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?



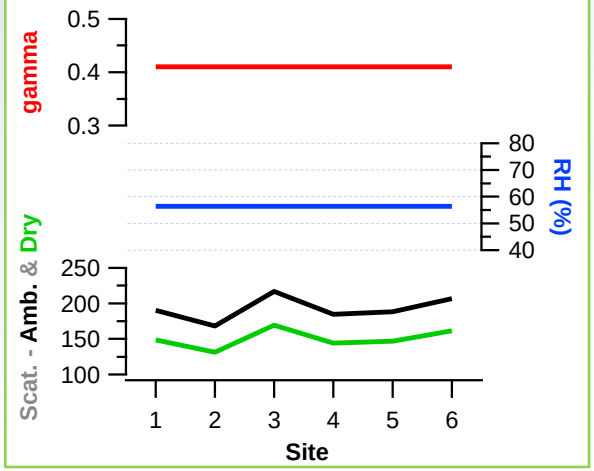
	Average	Std. Dev.	Rel. Std. Dev.
Gamma	0.408	0.07	2%
RH	56	8	14%
Dry Scat.	150	13	9%
Amb. Scat.	195	28	14%

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?

Measure variability due to:

1) Aerosol Loading

- Hold gamma & RH constant



Variability in ambient scattering (relative standard deviation, rsd):

9%

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?

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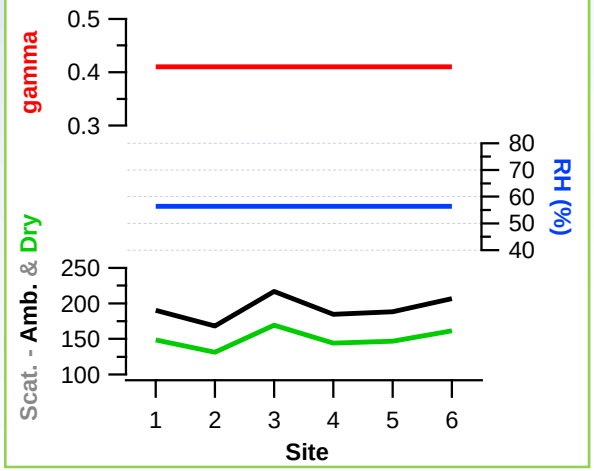
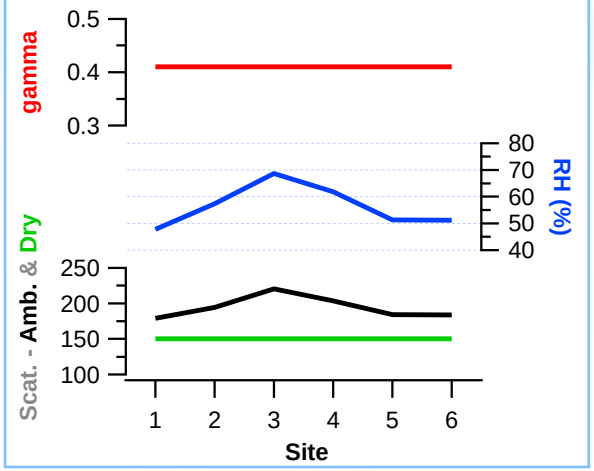


2) Relative Humidity

- Hold gamma & dry scattering constant



Variability in ambient scattering (relative standard deviation, rsd):

9% **8%**

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?

Measure variability due to:

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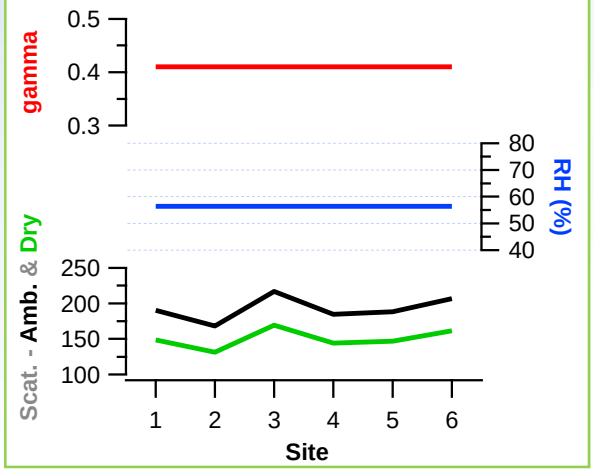
- Hold gamma & RH constant



2) Relative Humidity
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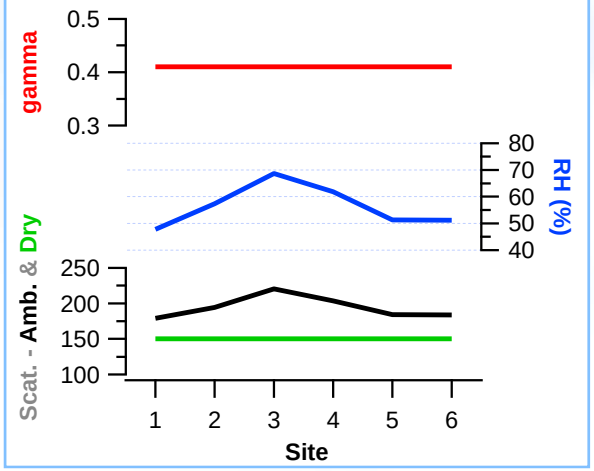
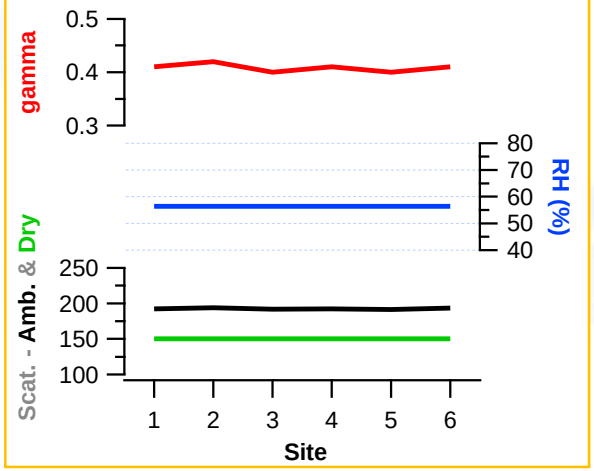


3) Composition
- Hold RH & dry scattering constant



Variability in ambient scattering (relative standard deviation, rsd):

9%	8%	0.4%

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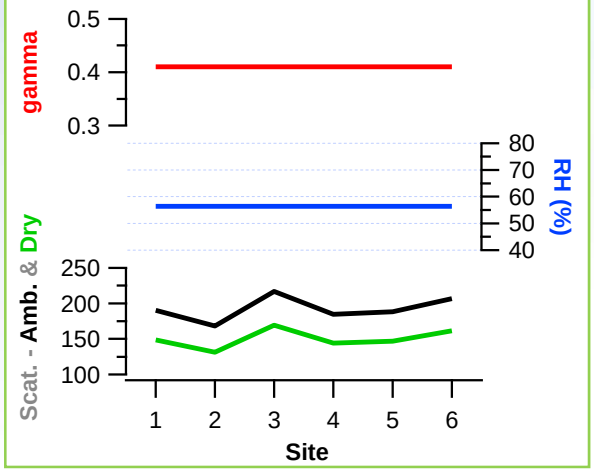
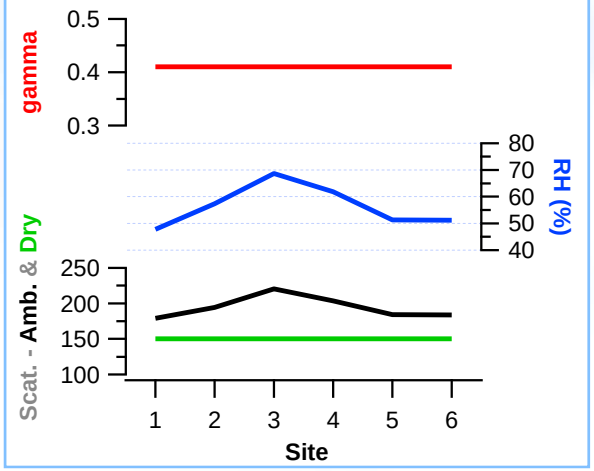


2) Relative Humidity
- Hold gamma & dry scattering constant



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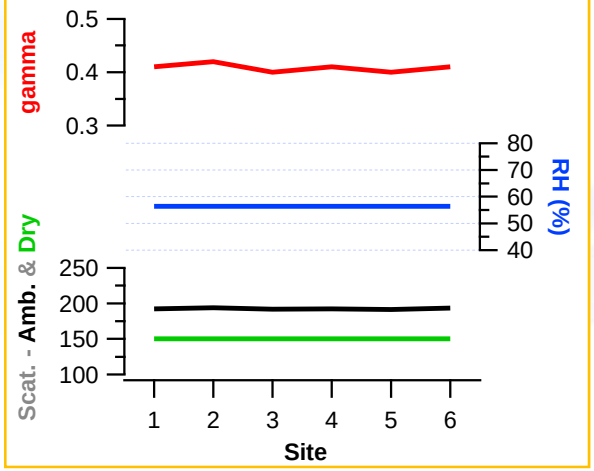

Variability in ambient scattering (relative standard deviation, rsd):

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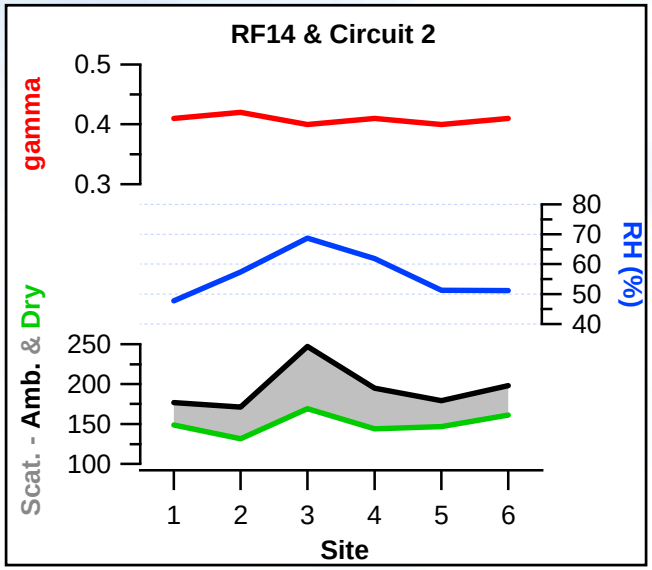
Relative Importance = $rsd_{dry} \times 100 / (rsd_{dry} + rsd_{RH} + rsd_{\gamma})$

51%	46%	3%

Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?



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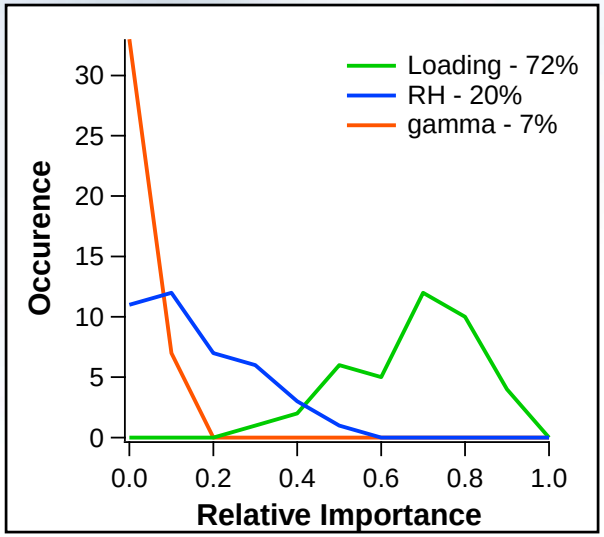
51% **46%** **3%**

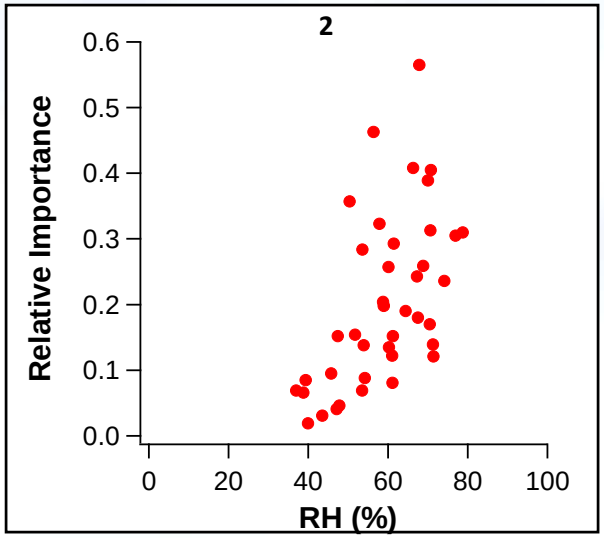
Variability Analysis: What Factors Have the Greatest Effect on Ambient Aerosol Scattering?

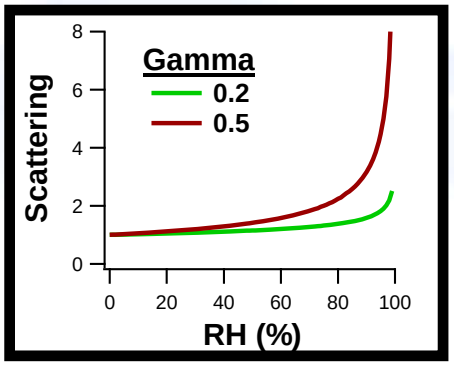
72% of the variability in aerosol scattering is due to variation in aerosol loading.

Additional 20% from variation in RH amongst the sites.

- Dependent on the RH due to non-linearity of aerosol hygroscopicity







Aerosol composition is a minor contributor to variation in ambient scattering/AOD

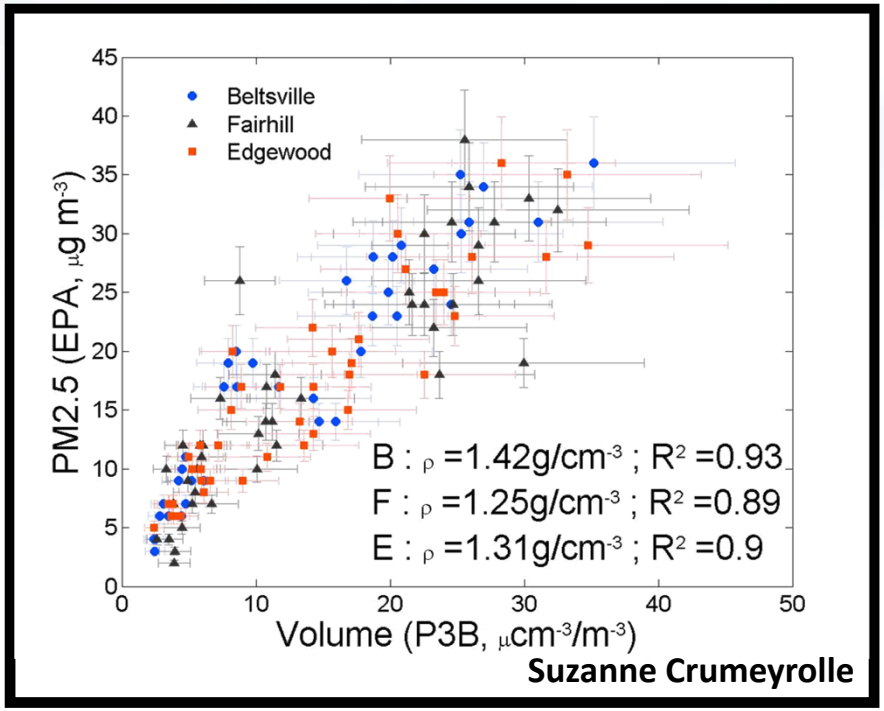
Ground and In Situ Aerosol Mass Comparison

PM2.5 measurements at 3 sites

- Measured by Beta-Attenuation Mass Monitor

Estimating mass from size distribution gives good agreement with the ground sites

- Particle density of 1.33 g/cm^3



Suzanne Crumeyrolle

Conclusions

Conclusions:

- Measurements in Baltimore during July 2011 sampled two distinct aerosol types:
  - Low loading days with 60% organic mass and smaller aerosols
  - High loading days with 60% ammonium sulfate and larger aerosols
- Aerosol hygroscopicity (γ) was dependent on the organic fraction of the aerosol
- Variability in RH amongst the sites accounts for 20% of the apparent variability in aerosol scattering
- In situ airborne measurements agreed well with ground-based measurements (density of 1.33)

Future Campaigns:

- January/February 2013 – Central Valley, CA
- September 2013 – Houston, TX
- Summer 2014 – location to be determined

Thanks:

- **NASA Earth Venture program through the Earth System Science Pathfinder Program Office**
- **Entire DISCOVER-AQ Team**

Aerosol Hygroscopicity

Gamma (γ) is dependent on composition

- $\gamma = 0.6 - 0.4 f_{POM}$
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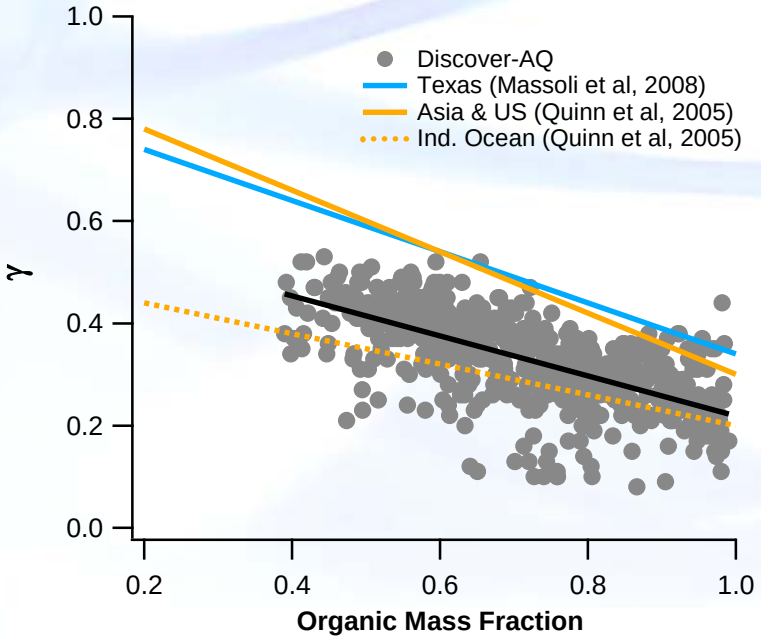
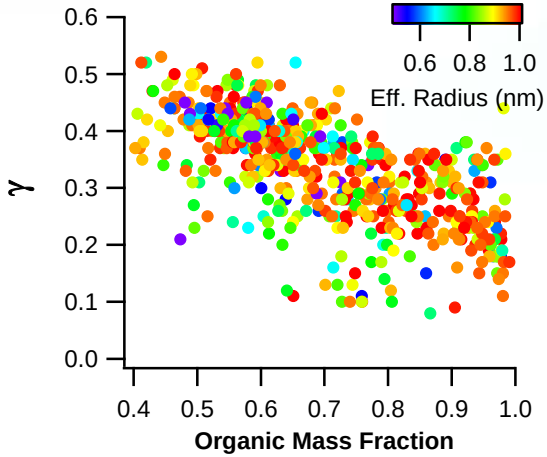
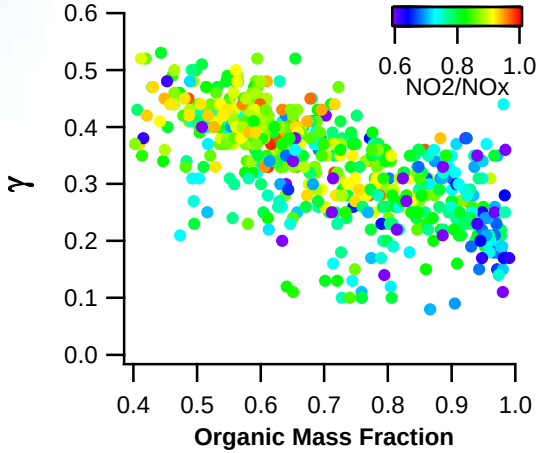






Remote and In Situ Scattering Comparison

Comparing LIDAR and in situ measurements

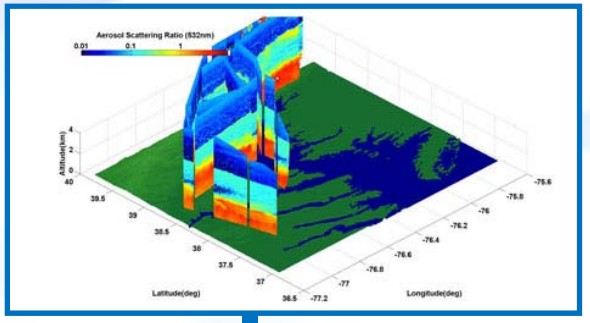
- **HSRL** – measures scattering at ambient RH
- **In Situ** – higher than ambient cabin temperature results in a reduced RH

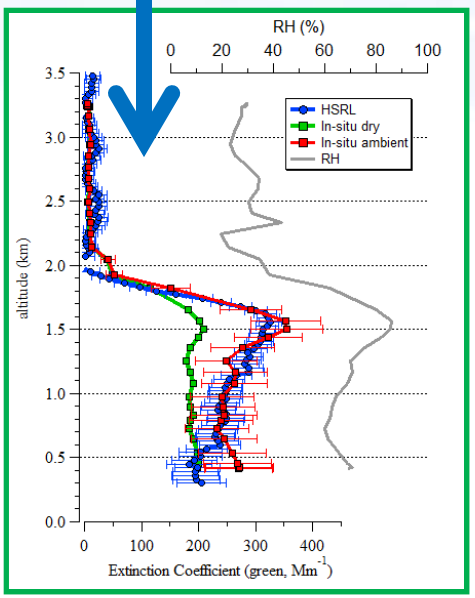
Good Correlation between HSRL and corrected in situ scattering

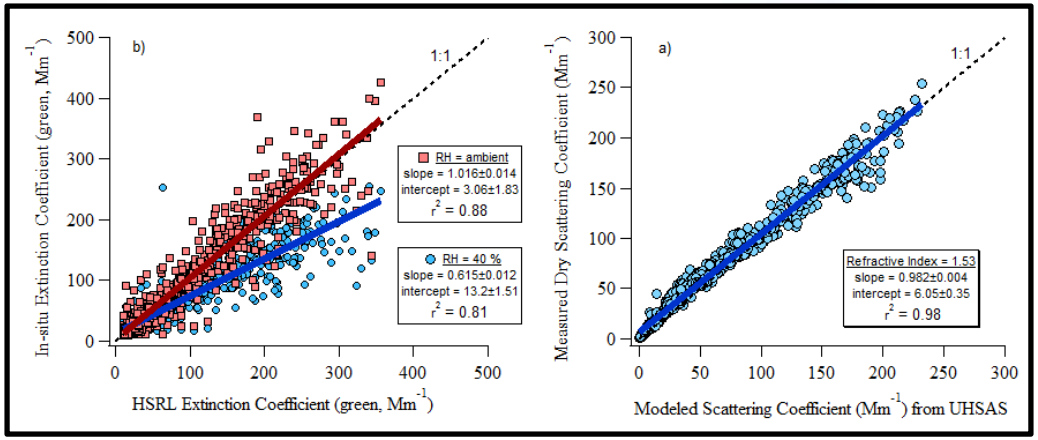
Dry scattering can also be compared to the aerosol size distribution data

- refractive index of 1.53(why) used

Accepted from publication in GRL (Ziemba et al.)







Mass Absorption Enhancement

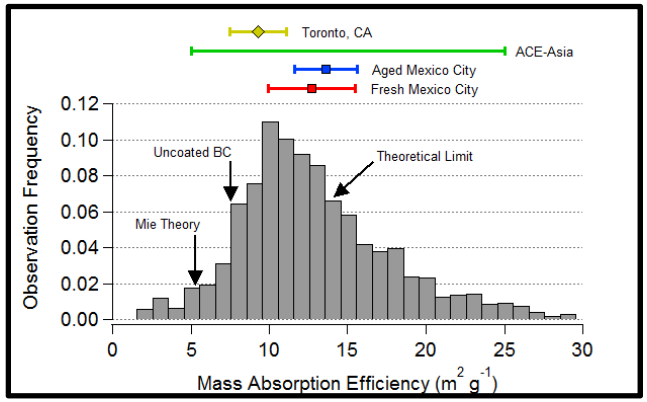
Black carbon mass is related to absorption via it's mass absorption efficiency (MAE) with units of m^2/g.

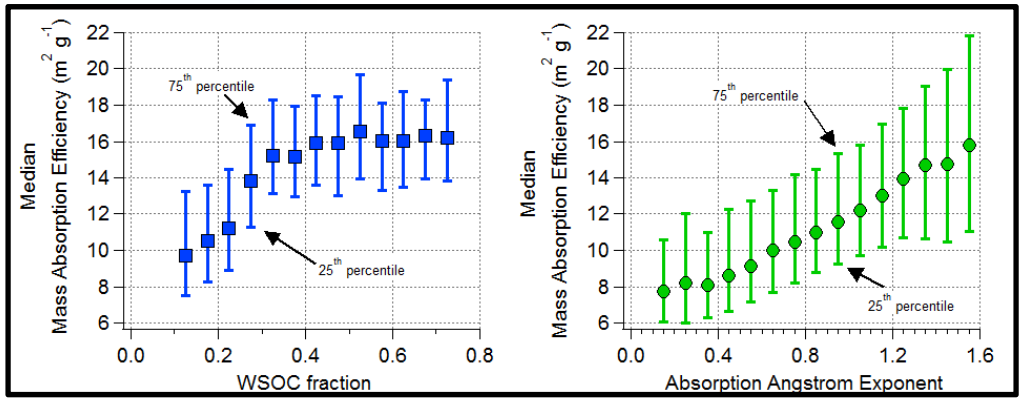
- Bare carbon = 7.5±1.2 (Bond, AS&T '06)
- Soot coating increases absorption by acting as a 'lens' for the incoming radiation

  ($MAE_{coated} = MAE_{bare} + abs_{coating}$)

Measured MAEs are considerably higher than MAE_{bare}

- Similar range as measured in other urban cities (Mexico City and Toronto)
- MAE increased with the WSOC fraction and absorption angstrom exponent (AE_{abs})
  - AE_{abs} uses solely optical measurements
  - Deriving MAE from AE_{abs} allows for a better understanding of aerosol absorption & composition based on solely-optical methods (remote sensors)





Diurnal Trends

Aerosol mass changes

- Increase in ammonium and sulfate due to increased photochemistry
- Decrease in organic and nitrate mass
- Black carbon modestly higher at midday

Total aerosol mass increased slightly causing an increase in aerosol scattering