Development of a Quantitative Accounting Framework for Black Carbon and Brown Carbon from Emissions Inventory to Impacts

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Motivation

- Emissions inventories and air quality models of light adsorbing carbon require parameterization of the radiative properties of emissions
- Current parameterizations of light absorbing carbon emissions do not address the range of variability within sources or control technologies
- Elemental carbon is not a good surrogate for light absorbing carbon for control strategy development nor assessment of control strategy implementation
  - May be OK if limited to absorption at 880 nm
- The light absorbing capacity of carbonaceous aerosol is not a conservative property from the point of emissions to atmosphere

Project Goals

- Overall Goal
  - Development of a quantitative framework for source-receptor relationships for light absorbing carbon and their associated wavelength dependent light absorptivity
- Key Objectives
  - Deconstruct emissions from sources of light absorbing carbon to elucidate the contribution of different emissions components to wavelength dependent absorption
  - Elucidate how the evolution of emissions in plumes impact wavelength dependent absorption
  - Integrate source apportionment models for aerosol components impacting light adsorption with wavelength dependent light absorption closure calculations

Project Strategy

- Source Testing
- Mie theory calculations for source emissions and deconstructed emissions
- Atmospheric measurements
- Mie theory calculations for atmospheric aerosols and deconstructed aerosols
- Develop a source apportionment framework that can address the optical evolution of aerosols and precursors

Source Testing

- Examine key sources of light absorbing carbon:
  - Mobile sources
    - Conventional CI and SI and Emerging Technologies
  - Biomass burning
    - Lab and Field Studies
  - Coal combustion
- Examine for each source
  - Role of process variables on emissions
  - Optical properties of the organic carbon
  - Optical properties of the elemental carbon
  - Impact of dilution
  - Impact of thermal stripping of organics
- Develop source specific light absorption closure models for measurement conditions and high dilution conditions

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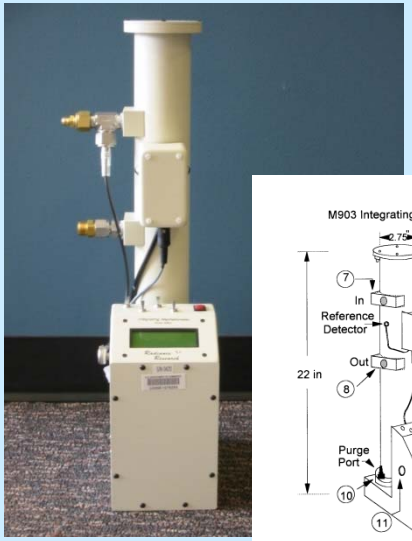
Atmospheric Measurements

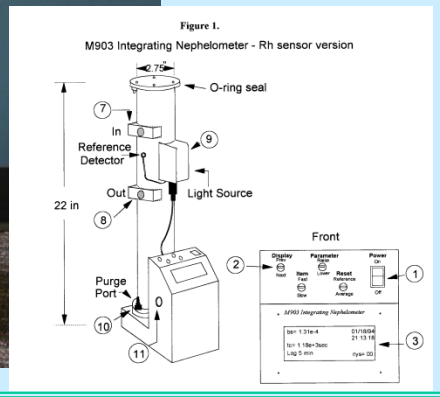
- Use sites where we have conducted source apportionment studies in the past and where historical record and optical measurements
  - Atlanta, Georgia
    - Near Roadway
  - Rural Alabama
    - SOA
  - India
    - Biomass and Trash Burning
    - Low Temperature Coal Combustion

Approach

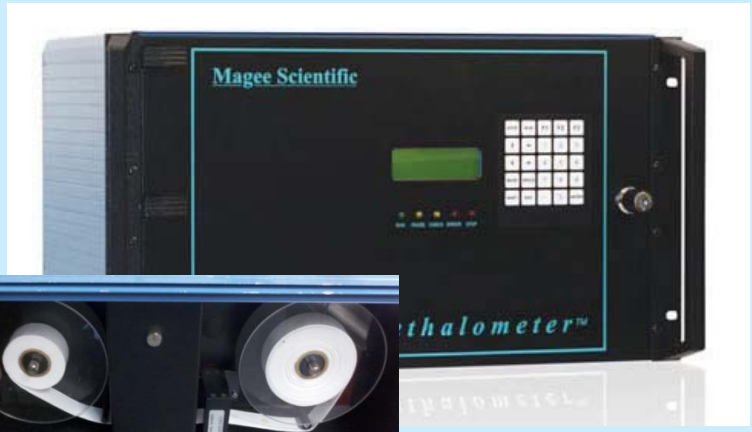
- Measure the optical properties under controlled conditions
  - Scattering and Absorption (multiple wavelengths)
- Measure physical-chemical properties
  - Size distribution, particle shape, chemical composition
- Segregate components of aerosols
  - Thermal Denuder, WS and Organic solvent atomization
- Correct absorption artifacts and compare optical properties of aerosol components

Methods



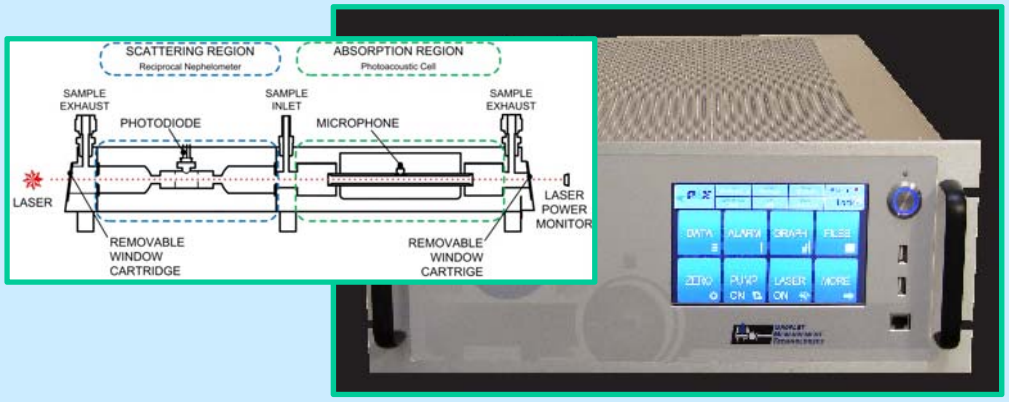


Radiance Research Nephelometer



Magee Scientific AE31 7-channel Aethalometer

TSI Scanning Mobility Particle Sizer/ Electrostatic classifier



DMT PAX 532: Photoacoustic Extictionometer

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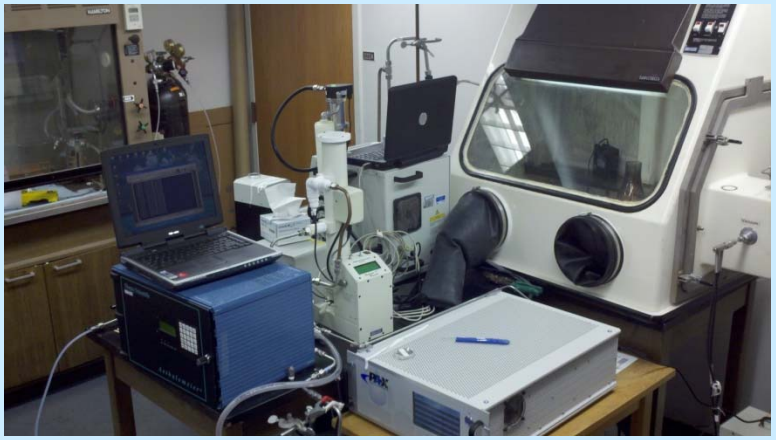
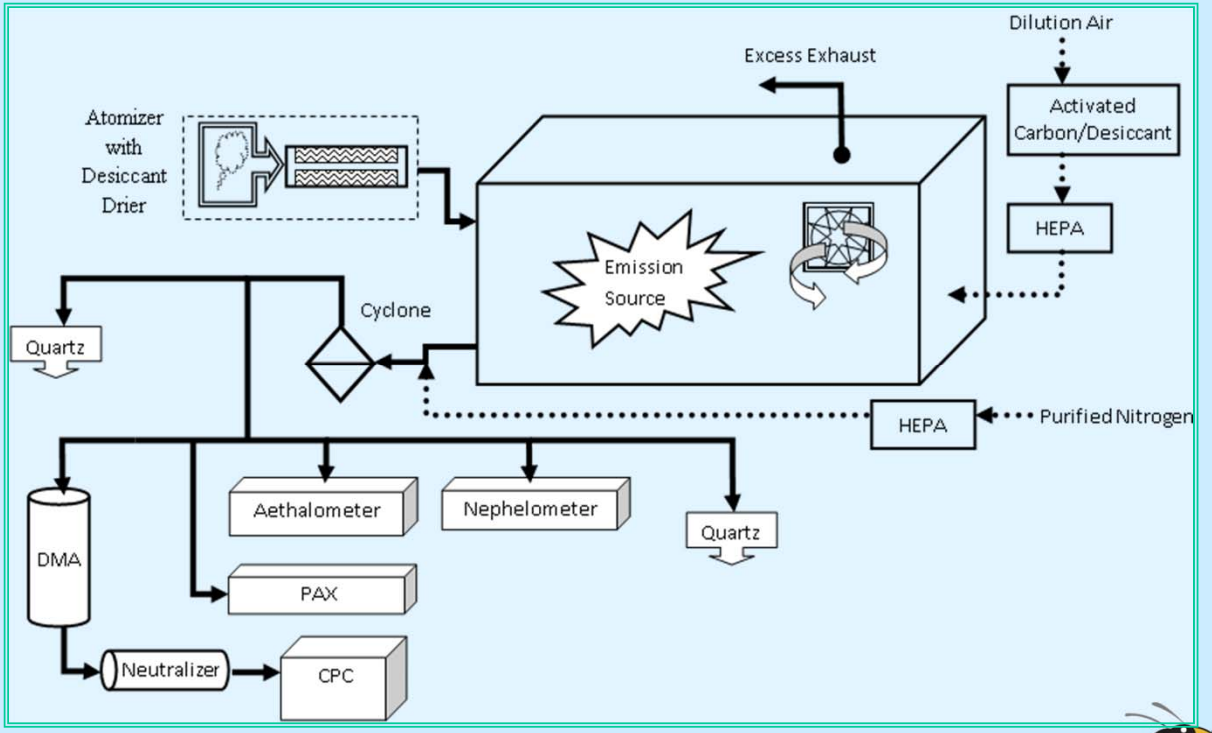
Methods





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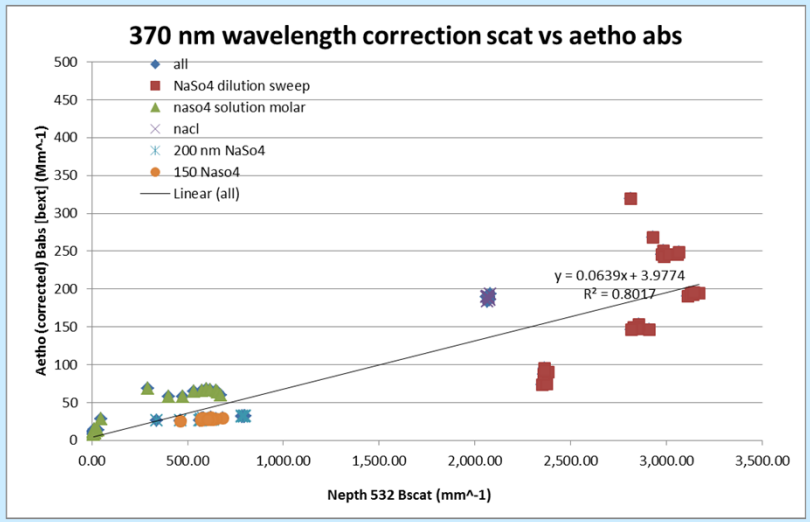
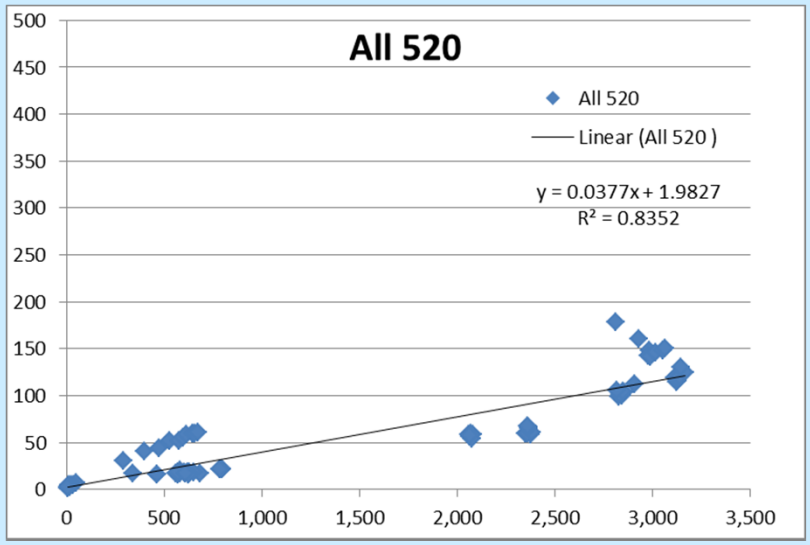
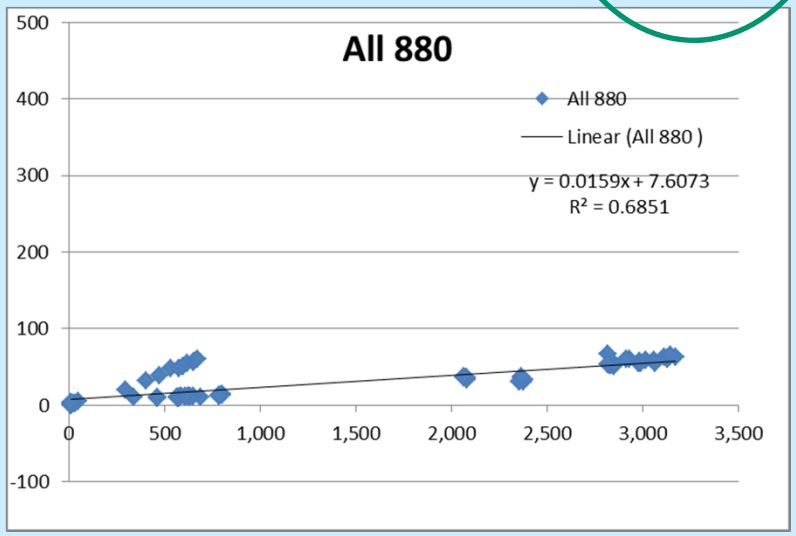
Attenuation by Non-Absorbing Aerosols



Absorption vs Scattering: Scattering Artifact correction

Wavelengths	slope (m)	Intercept (b)	R^2	slope forced through zero (m')
370	**0.064**	**3.977**	0.801747	**0.066**
470	**0.049**	**-0.909**	0.816126	**0.048**
520	**0.038**	**1.983**	0.835205	**0.039**
590	**0.030**	**2.919**	0.801001	**0.032**
660	**0.027**	**3.207**	0.84761	**0.028**
880	**0.016**	**7.607**	0.685092	**0.019**
950	**0.013**	**7.410**	0.651662	**0.016**





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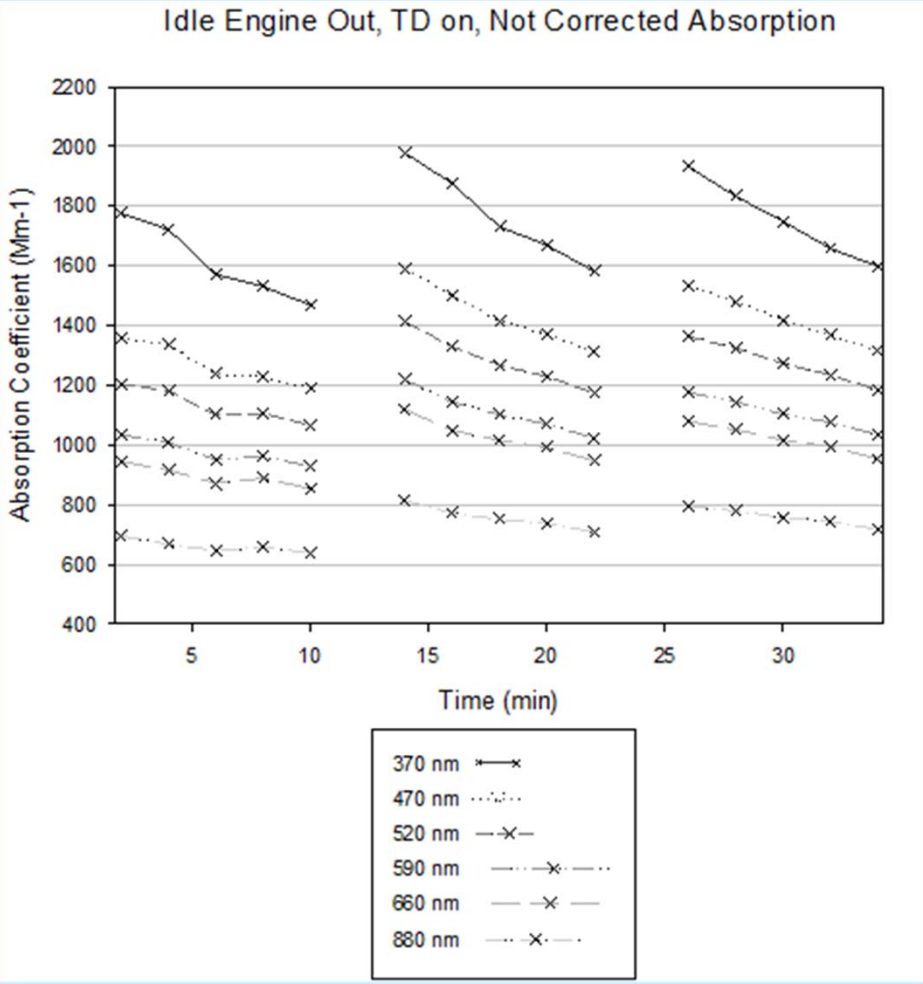
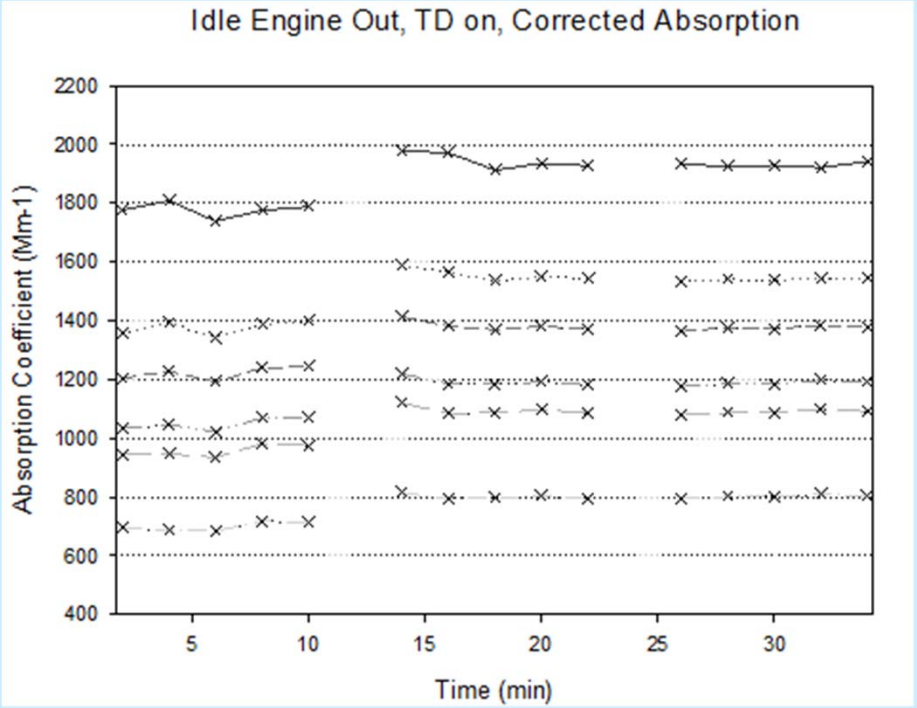
Multiple Wavelength Absorption Correction





- Test run at steady-state
- Scattering correction is not significant for engine out emissions
- Loading correction is wavelength specific

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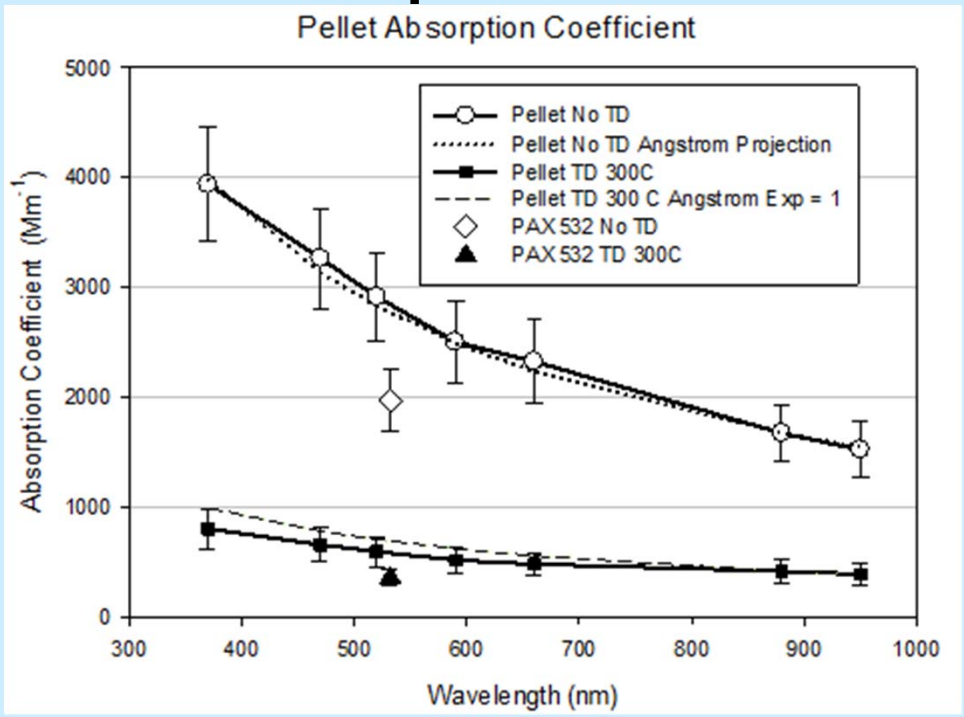
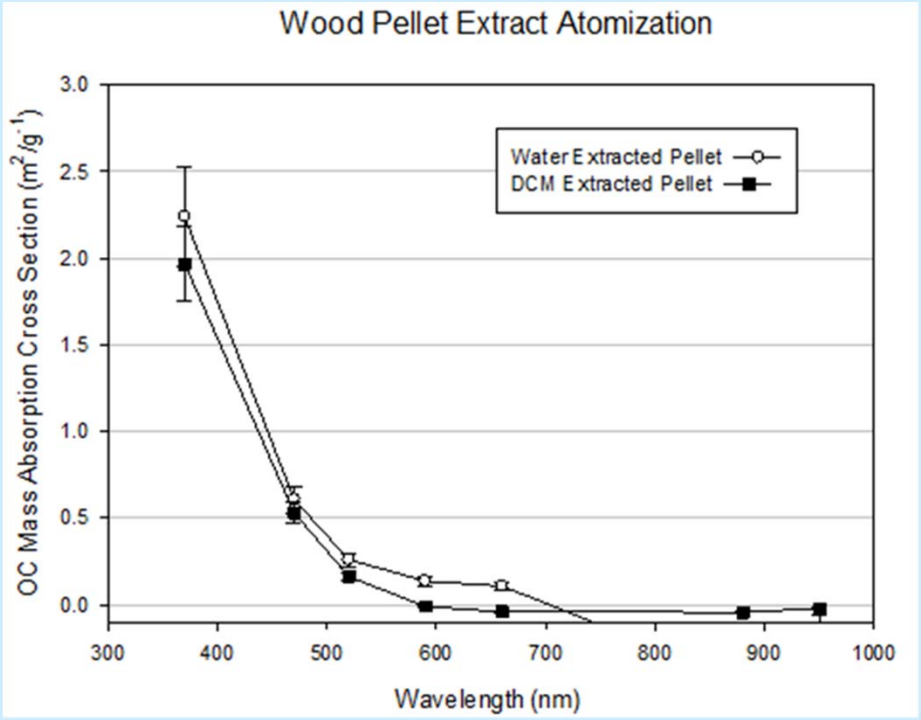
Example of Wood Pellets





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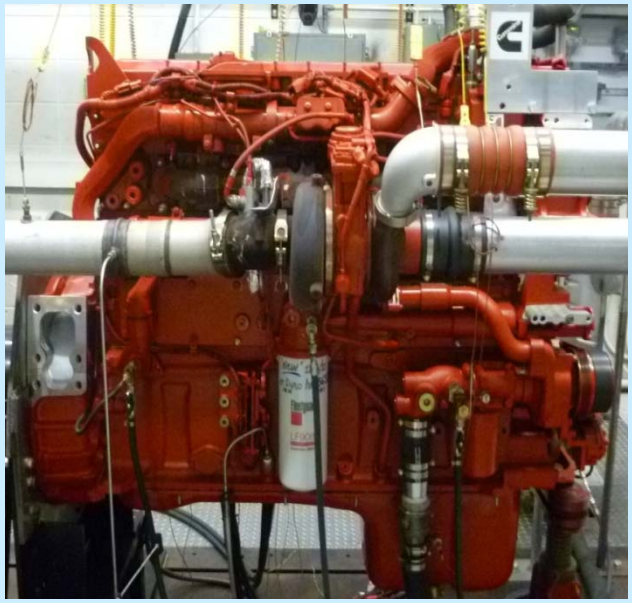
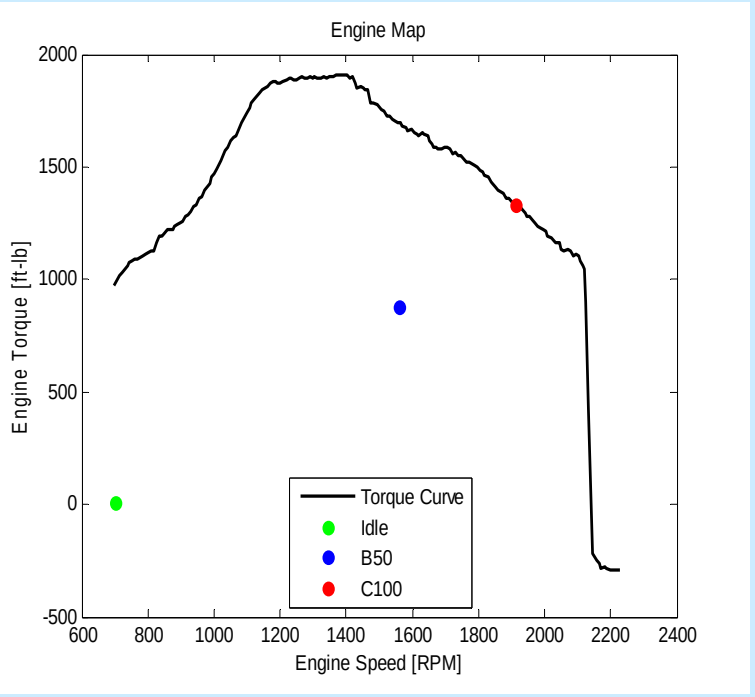
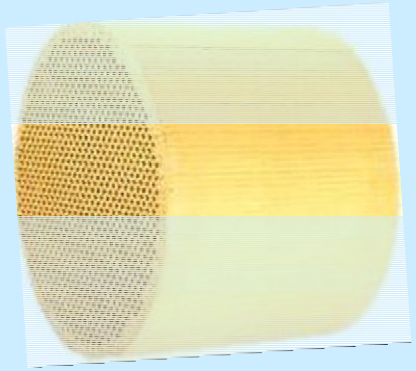
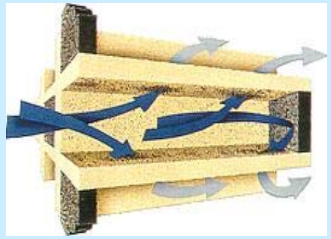
Experimental Setup



Model	2010, Cummins ISX15 – 500
Emission Certification	EPA 2010, CARB 2010
Type	4-stroke cycle
Cylinder Configuration	In-line 6
Bore and Stroke	137 mm x 169 mm
Compression Ratio	17.2:1
Aspiration	Turbocharged & Charge Air Cooled
Displacement	14.9 L
Rated Power & Rated Speed	373 kW & 1800 RPM
Peak Torque	2508 N-m at 1200 RPM
Fuel System	Cummins XPI
EGR System	Cooled High Pressure







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Emissions Testing Lab

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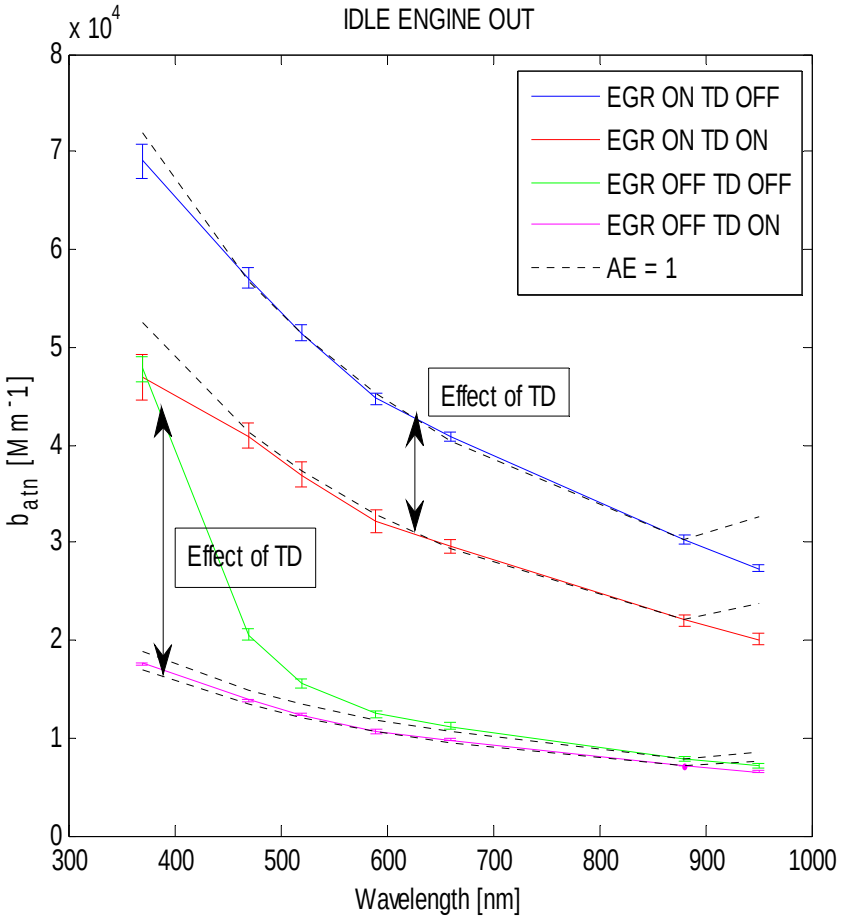
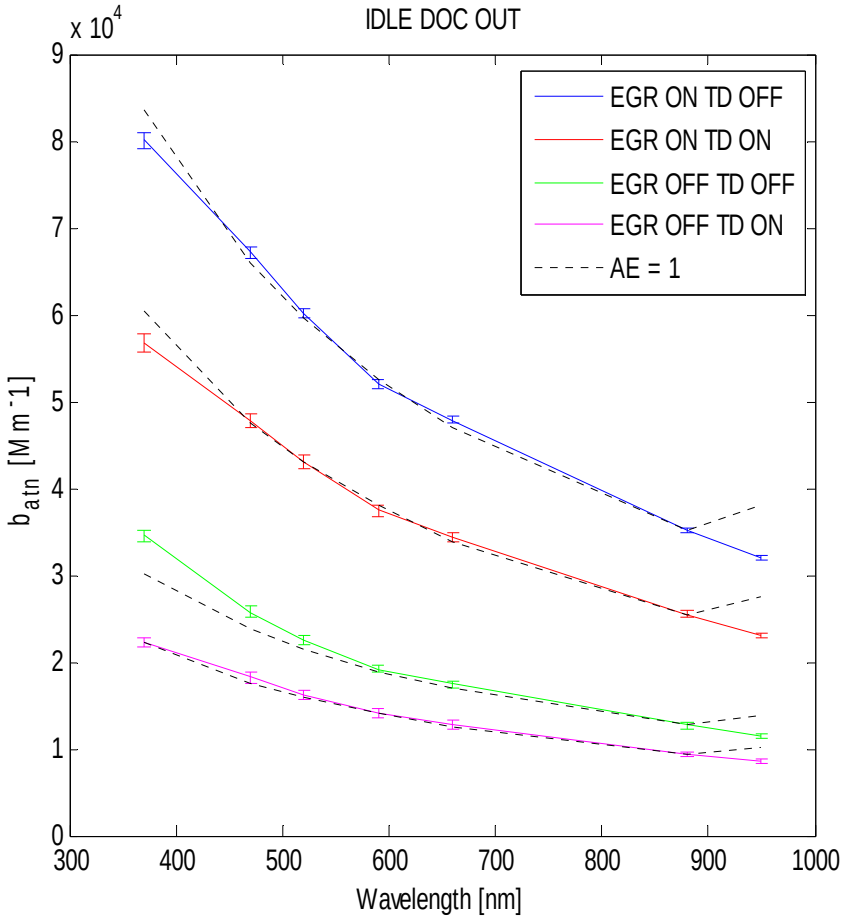
BrC Plots





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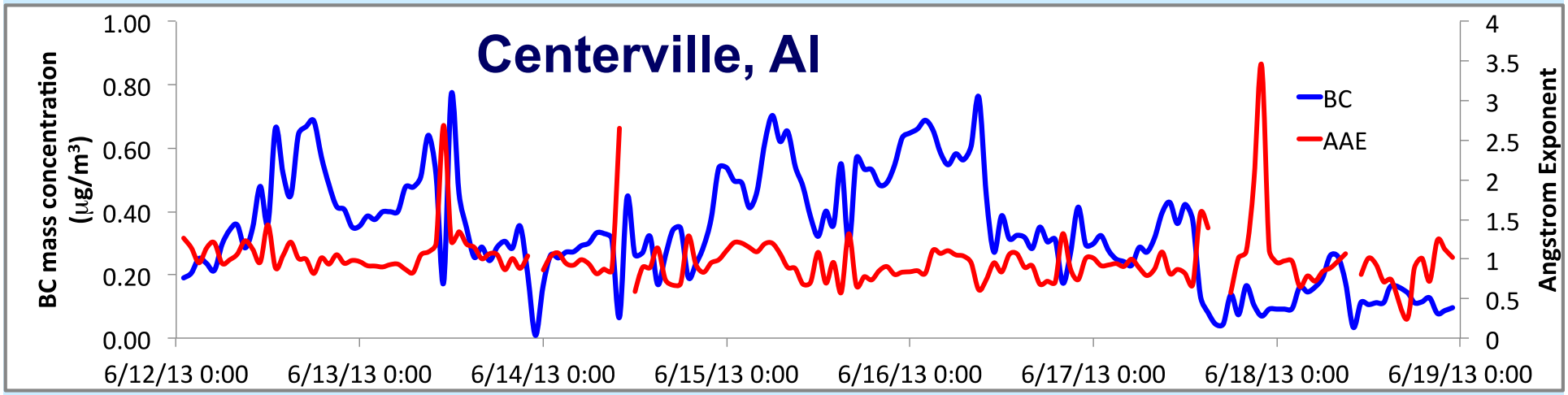
Atmospheric Sampling: Objectives

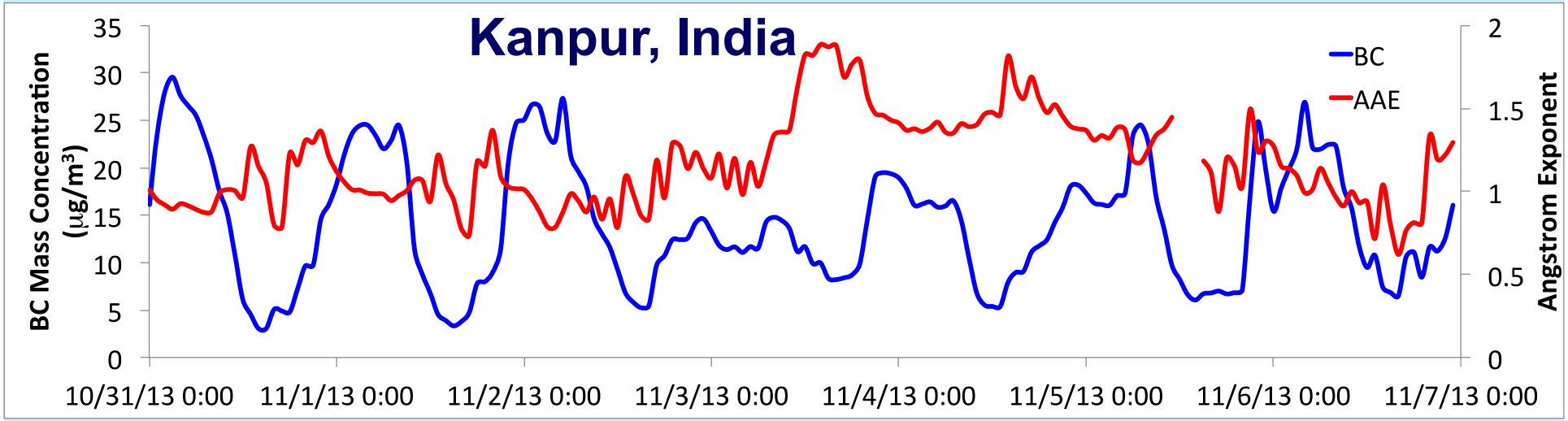
- Conduct field measurements at a variety of sites dominated by various sources of Black Carbon (BC) and Brown Carbon (BrC)
- Determine relative fraction of light absorption by BC and BrC
- Determine sources of BC and BrC
- Develop simple parameterizations for influence of aging on the light absorbing properties of aerosols

Approach: Specifics

Parameter	Instrument	Dates	Objective
Real-Time Continuous			
σ_{ap} (λ)	Magee Aethalometer, PAX	4-1 Month Periods	Compare with Mie Theory Light Absorption Estimates from MOUDI 1
σ_{sp} (λ)	Radiance Research Nephelometer	--	Compare with Mie Theory Estimates from MOUDI
Time-Integrated Sampling			
EC/OC, Trace Organics, WSOC, $Abs(\lambda)_{solvent}$, $Abs(\lambda)_{water}$	HiVol Filter sampler	4-1 Month Periods	Source apportionment, RI Estimates for Mie Theory, Solvents Extracts for Aerosolization Experiments
EC/OC, WSOC, $Abs(\lambda)_{solvent}$, $Abs(\lambda)_{water}$	MOUDI 1	4-1 Month Periods	Estimation of σ_{ap} (λ) as function of size for both water and solvent extracts and BC
Mass, Ions	MOUDI 2	4-1 Month Periods	Estimation of σ_{sp} (λ) as a function of size

Black Carbon (BC) and Angstrom Absorption Exponent (AAE) in rural US and India





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Trash/Refuse Burning: A Source of Brown Carbon

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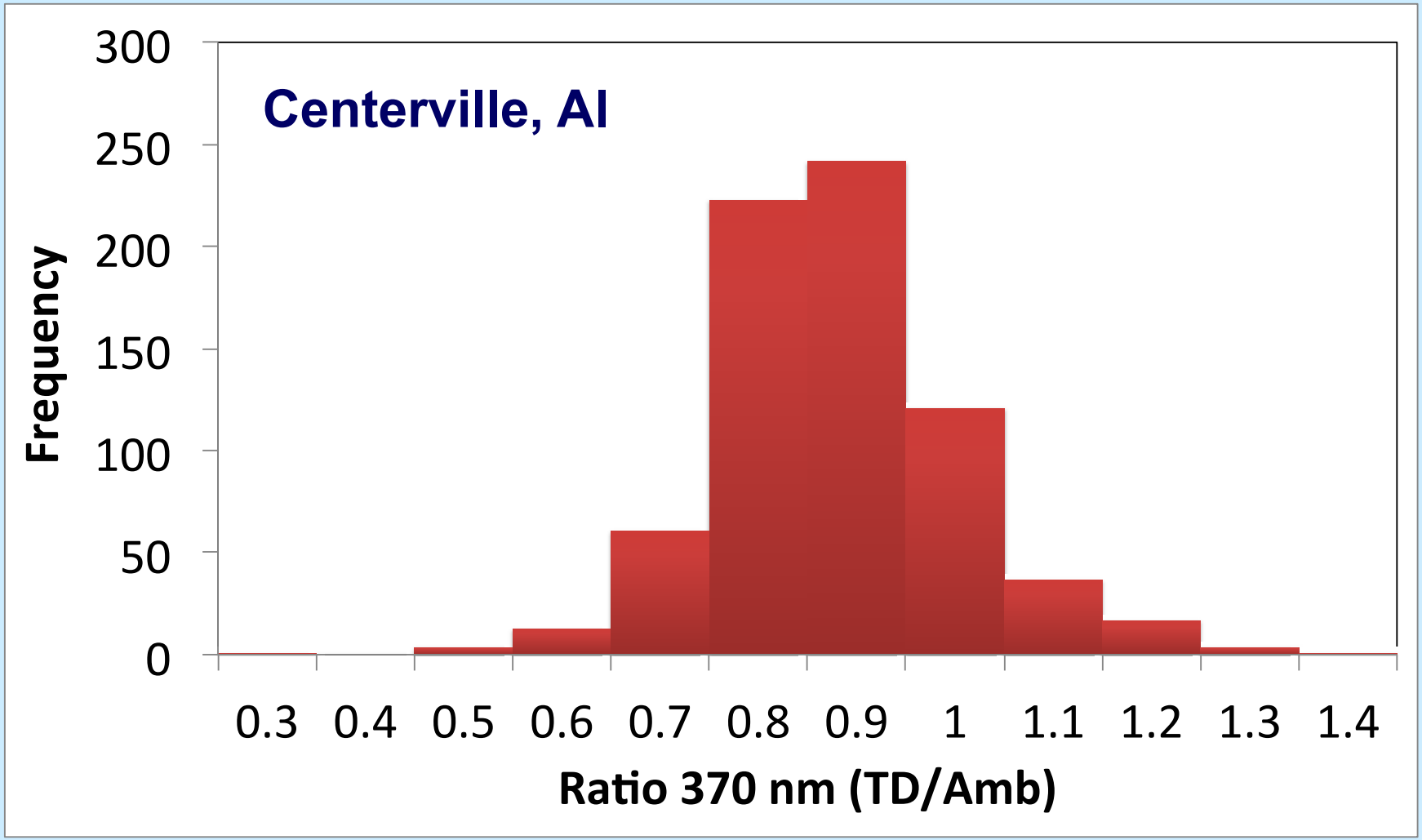
Ratio of light absorption for denuder (200°C) to ambient air



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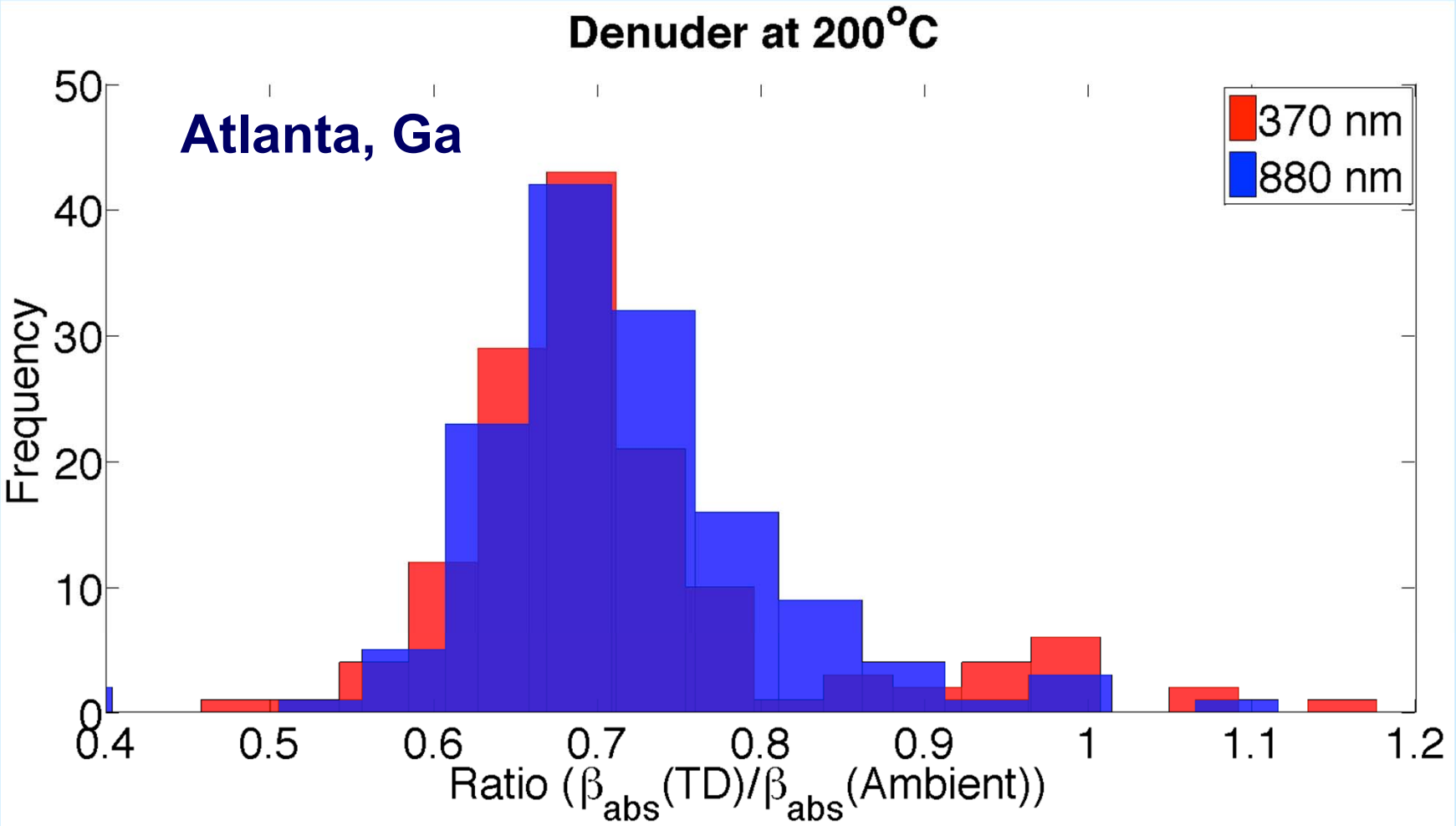

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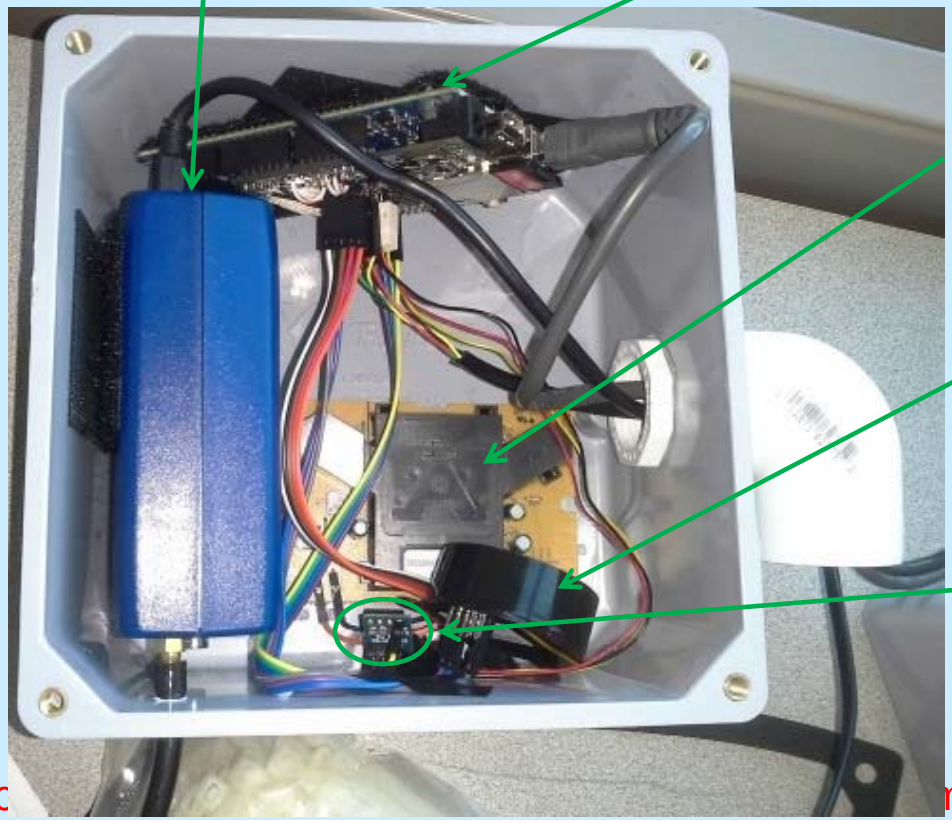
Low Cost Sensor Networks



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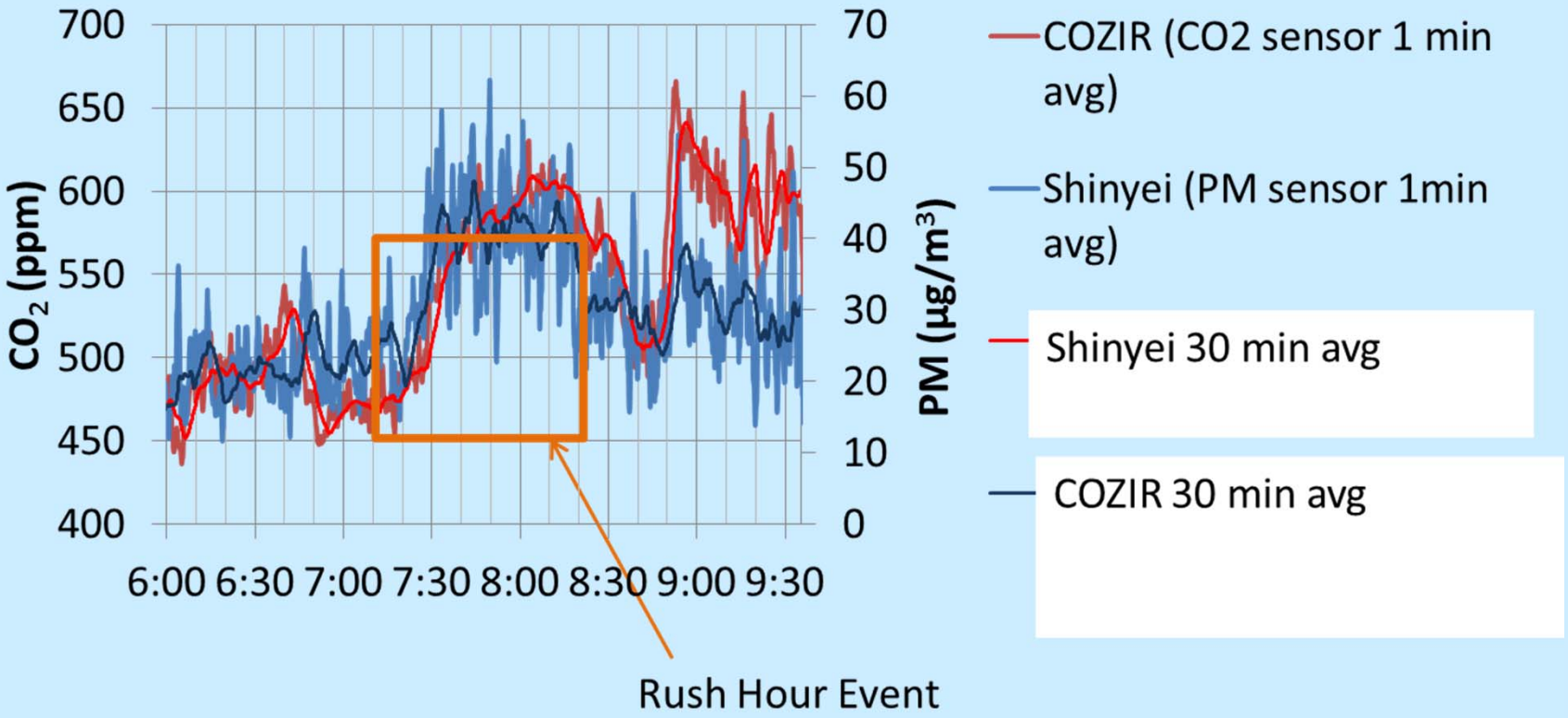
Atlanta Freeway PM Emission Factor Estimate



Rough Emissions Factor = ΔPM/ΔCO2
= 0.079µg m^{-3} PM/ppmCO2
= 0.39 g PM/kg fuel

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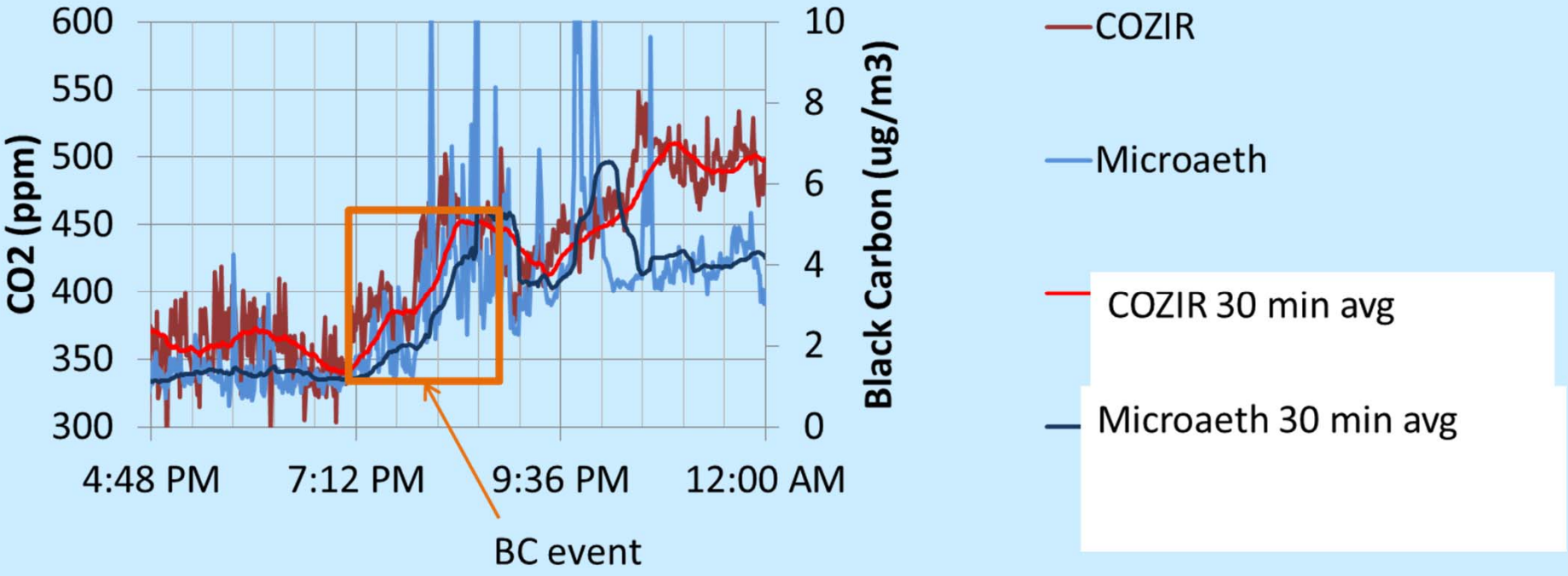
Atlanta Freeway BC Emission Factor Estimate



Rough Emissions Factor = ΔBC/ΔCO2
= 0.044 μg m^{-3} BC/ppmCO2
= 75 mg BC/kg fuel

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Ongoing Efforts

- Source Testing
  - Appling methodology to other source of concern: real world biomass, residential coal
- Atmospheric Sampling
  - Water and methanol extractions of size-resolved BC and BrC samples
  - Extraction of hivol samples to determine optical properties and sources of light absorption
- Publications