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

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Grant # [blank]

Framework for Black Carbon and Brown Carbon from Emissions to Impacts
EPA STAR Grant R83503901
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

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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 Extinctionometer

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Methods

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Attenuation by Non-Absorbing Aerosols

Absorption vs Scattering: Scattering Artifact correction

<table>
<thead>
<tr>
<th>Wavelengths</th>
<th>slope (m)</th>
<th>Intercept (b)</th>
<th>R^2</th>
<th>slope forced through zero (m')</th>
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</thead>
<tbody>
<tr>
<td>370</td>
<td>0.064</td>
<td>3.977</td>
<td>0.801747</td>
<td>0.066</td>
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<tr>
<td>470</td>
<td>0.049</td>
<td>-0.909</td>
<td>0.816126</td>
<td>0.048</td>
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<tr>
<td>520</td>
<td>0.038</td>
<td>1.983</td>
<td>0.835205</td>
<td>0.039</td>
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<tr>
<td>590</td>
<td>0.030</td>
<td>2.919</td>
<td>0.801001</td>
<td>0.032</td>
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<tr>
<td>660</td>
<td>0.027</td>
<td>3.207</td>
<td>0.84751</td>
<td>0.028</td>
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<tr>
<td>880</td>
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<td>7.607</td>
<td>0.685092</td>
<td>0.019</td>
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<tr>
<td>950</td>
<td>0.013</td>
<td>7.410</td>
<td>0.651662</td>
<td>0.016</td>
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</tbody>
</table>

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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

Pellet Absorption Coefficient

Wood Pellet Extract Atomization

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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

Effect of TD

Effect of TD

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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

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

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

Centerville, Al

Kanpur, 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

Centerville, Al

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

Denuder at 200°C

Atlanta, Ga
Low Cost Sensor Networks

- microAet h-Black Carbon
- Arduino microcontroller
- PM sensor
- CO2 Sensor
- T, RH

Framework for Black Carbon

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Rough Emissions Factor = $\frac{\Delta \text{PM}}{\Delta \text{CO}_2}$

- $0.079 \mu g \ m^{-3} \ \text{PM/ppmCO}_2$
- $0.39 \ g \ \text{PM/kg fuel}$
Atlanta Freeway BC Emission Factor Estimate

Rough Emissions Factor = $\Delta$BC/$\Delta$CO2

= 0.044 $\mu$g m$^{-3}$ BC/ppmCO2

= 75 mg BC/kg fuel

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Ongoing Efforts

• Source Testing
  – Applying 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

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