Retrieval of Black Carbon Concentration from the Aerosol Robotics Network (AERONET)

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Oleg Dubovik, ELICO, Université du Littoral Côte d’Opale
Brent Holben, NASA GSFC

Worldwide black carbon concentration measurements are needed to assess the efficacy of the carbon emissions inventory and transport model output. This requires long-term measurements in many regions, as model success in one region or season does not apply to all regions and seasons. AERONET is an automated network of more than 180 surface radiometers located throughout the world. The sky radiance measurements obtained by AERONET are inverted to provide column-averaged aerosol refractive indices and size distributions for the AERONET database, which we use to derive column-averaged black carbon concentrations and specific absorptions that are constrained by the measured radiation field. This provides a link between AERONET sky radiance measurements and the elemental carbon concentration of transport models without the need for an optics module in the transport model. Knowledge of both the black carbon concentration and aerosol absorption optical depth (i.e., input and output of the optics module) will enable improvements to the transport model optics module.
Black Carbon Concentration from Worldwide Aerosol Robotic Network (AERONET) Measurements

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Outline

- Motivation
- AERONET product
- Maxwell Garnett effective medium approximation
- Description of black carbon retrieval and results
- Validation and sensitivity study
Modeling Global Black Carbon Absorption

Carbon Emissions Inventory

Dispersion, Transport, and Removal

BC Mass; $M \text{ (g m}^{-3}\text{)}$

Optics Module; $\alpha \text{ (m}^2\text{g}^{-1}\text{)}$

Absorption; $\tau_{\text{abs}} = \alpha M_i \Delta Z_i$

Uncertainty factor > 2
Modeling Global Black Carbon Absorption

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External Mixture

Tuning factor: 2 to 4
(Sato et al., 2003)
Modeling Global Black Carbon Absorption

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Modeling Global Black Carbon Absorption

- **Carbon Emissions Inventory**
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- **Absorption;** \( \tau_{\text{abs}} = \alpha M_i \Delta Z_i \)
- **Uncertainty factor > 2**
- **Tuning factor: 2 to 4** (Sato et al., 2003)

- **This Study**
- **Internal Mixture**
- **AERONET**

- **Park et al. (2003)**
- **External Mixture**
- **15%**
Based upon almucantar sky radiance scan
Avg residual radiance errors < 5%, 21 angles

Provides columnar size distribution
at 22 radii from 0.05 to 15 µm

Complex refractive index at 4 wavelengths
\( n(\lambda), k(\lambda) \)

Internal mixture

Cloud-screening: temporal, spatial, and symmetry constraints
Maxwell Garnett effective medium approximation

Maxwell Garnett refractive index:

\[ m_{MG}(m_{host}, m_j, f_j) \]

\[ f_j = \text{inclusion volume fraction} \]

Assumptions:
(small, spherical inclusions)

Electron microscope photograph

Discrete dipole model

Martins et al., JGR, 1998
Maxwell Garnett effective medium approximation

Maxwell Garnett refractive index:

\[ m_{MG}(m_{host}, m_j, f_j) \]

\( f_j = \text{inclusion volume fraction} \)

Assumptions:
(small, spherical inclusions)

52% Black carbon, monomer radii up to 57 nm, particle radii 0.05-0.4 \( \mu \text{m} \)

Maxwell Garnett specific absorption consistent with discrete dipole model to within \( \sim 10\% \).
Black carbon content from AERONET retrievals

\[ \chi^2 = \sum_{l=1}^{4 \text{ wvlns}} \left( \frac{m_l^{rtrv} - m_l^{mix}}{m_l^{rtrv}} \right)^2 \Rightarrow 0 \]
Yearly averaged black carbon concentration

- N. Amer.
- Europe
- Asia
- S. Amer.
- Africa
- Atlantic
- Pacific

BC concentration (mg m$^{-2}$)

Continental Sites

Biomass Burning

Atlantic and Pacific Islands

circles - 2000
squares - 2001
Black Carbon Specific Absorption

\[ \alpha = \frac{\tau_a}{[BC]} = \frac{\tau_a \left( m_{mix} \left( f_{BC} \right) \right)}{f_{BC} \rho_{BC} \int \frac{dV}{d \ln r} d \ln r} \]

- $\tau_a$ absorption AOT
- $f$ component fraction
- $m$ refractive index
- $\rho_{BC}$ density of black carbon
BC Specific Absorption for Nine Nondust Climatologies
BC Specific Absorption for Nine Nondust Climatologies

Also reported by Petzold et al., 1997; Neusüß et al., 2002
GSFC

45 AERONET sites  2000 - 2001

GSFC

\( \lambda = 550 \text{ nm} \)
Yearly-averaged black carbon specific absorption

$\lambda = 550 \text{ nm}$

2000 -- circles
2001 -- squares

N. Amer.
Europe
Asia
S. Amer.
Africa
Atlantic
Pacific
Pseudo-validation

Model Comparison to RSS irradiance at ARM SGP site

Description of RSS (Rotating Shadowband Spectroradiometer)

- Spectral irradiance, 0.36-1.1 µm
- 193 measurements coincident w/ AERONET retrievals in 2000-2002

Model Atmosphere

- Correlated-k distribution for gas absorption (Kato et al., 1999)
- Microwave radiometer and other instruments: P(Z), T(Z), H$_2$O$_v$(Z)
- Molecular extinction and O$_2$ absorption scaled to P(Z)
- AERONET size distribution, constrained to 2-km boundary layer
- Minimum $\chi^2$ fit for AERONET aerosol refractive index
Model comparison with RSS irradiance

Kato et al., 1999 CKD; 0.35-1.05 µm,
Sensitivity of retrieved black carbon to choice of host aerosol

![Graph showing the sensitivity of retrieved black carbon to choice of host aerosol. The graph plots the mixture imaginary refractive index against black carbon volume fraction. Different markers and lines represent different host aerosols, with values for the refractive index of the host aerosol and the volume average indicated. The wavelength is given as 0.55 μm.]
III. Sensitivity to Assumptions

<table>
<thead>
<tr>
<th></th>
<th>[BC] Error (%)</th>
<th>Specific Absorption Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nonwater host</td>
<td>+15</td>
<td>-15</td>
</tr>
<tr>
<td>Coarse mode</td>
<td>??</td>
<td>??</td>
</tr>
<tr>
<td>OC</td>
<td>+10</td>
<td>-10</td>
</tr>
<tr>
<td>Maxwell Garnett</td>
<td>+/- 10</td>
<td>-/+ 10</td>
</tr>
<tr>
<td>BC density</td>
<td>+/- 5</td>
<td>-/+ 5</td>
</tr>
<tr>
<td></td>
<td>-15 to +40</td>
<td>-40 to +15</td>
</tr>
</tbody>
</table>

- Improvement over factor of 2+ uncertainty in BC emissions inventories
- Some cancellation of errors is likely
  Avg specific absorption for 19,591 retrievals is 9.9 m²g⁻¹
  Accurately calculates surface radiation
CONCLUSIONS

- BC concentrations and specific absorptions at 46 AERONET sites
- Results look reasonable
- “Pseudo-validated” with independent radiation measurements
- Internally-mixed BC absorption is sensitive to the details of the size distribution and the fraction of black carbon in the mixture
- Volume averaging for internal mixtures produces refractive indices that are too high; Maxwell Garnett equations are easily parameterized.

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Appendix
Black carbon is not a sphere

Aggregate soot at Sagres, Portugal;
Li et al., JGR (2003)
Black carbon is not a sphere

Aggregate absorption can be modeled as a loose collection of spheres to within 10-14% 
(Mulholland et al, 1994; Fuller, 1995)

Aggregate soot at Sagres, Portugal; 
Li et al., JGR (2003)
Comparison with Sunset Labs EC/OC Analyzer at Baltimore EPA Supersite

EPA Data courtesy of P. Hopke and M. Adam

\[ y = 0.6973x - 0.13 \]

\[ R^2 = 0.7971 \]
Sevilleta, New Mexico

Day of year

AOT (0.44 μm)

Day 132
Central America smoke appears in the news

Days 302-308
Hurricane Mitch ravages C. Amer.
Sensitivity of specific absorption (550 nm) to BC refractive index.

<table>
<thead>
<tr>
<th>Material</th>
<th>Refractive Index</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soot G</td>
<td>2 - 1i</td>
<td>2</td>
</tr>
<tr>
<td>Graphite</td>
<td>2.67 - 1.34i</td>
<td>2.26</td>
</tr>
<tr>
<td>OPAC</td>
<td>1.75 - 0.44i</td>
<td>2</td>
</tr>
</tbody>
</table>
Sensitivity of specific absorption (550 nm) to BC refractive index
Refractive index extrapolation; COVE, November 1, 1999, 17:48 GMT

- □ AERONET
- ○ χ-squared fit; 27% ammonium sulfate, 1.3 % soot by volume

Aerosol optical depth (0.440 μm) = 0.23
GSFC and WASH1 comparison
21 km between sites

- AERONET BC -- instantaneous, clear sky
- IMPROVE EC, fine mode -- 24-hr avg

2000
Internal and External Aerosol Mixtures

Internal mixing with layered structure:
- Non-absorbing shell (organics, sulfates, etc.)
- Black carbon core

External mixing:
- Black carbon
- Non-absorbing particles

Internal mixing in soot aggregates:
- Open soot cluster
- Closed soot cluster

Internal mixture

Martins et al., JGR, 1998
Fraction of aerosol forcing at RSS wavelengths
Model comparison to principle plane radiances

- Principle plane represents an independent measurement
- Homogenous troposphere of aerosols and molecules
- Ozone absorption in stratosphere using TOMS dataset

Compare average calculated radiance to measurements at four scanning wavelengths (0.44, 0.67, 0.87, 1.02 µm)

\[
\text{Error} = \frac{1}{4} \sum_{j=1}^{4} \frac{I_{j}^{\text{calc}} - I_{j}^{\text{meas}}}{I_{j}^{\text{meas}}}
\]
Graphite

Untreated
Heat treated (2500–2800 °C)

\[ \rho, \text{ g/cm}^3 \]

\[ d(002), \text{ Å} \]
Irradiance errors in the k-distribution bands
Comparison with RSS at ARM CF, 193 retrievals in 2000-2002.
Model includes aerosols, ozone, water vapor and $O_2$
Figure 8. High-resolution TEM image of soot from Sagres showing the discontinuous onion-like structure of graphitic layers.
Relative Humidity Effects on Soot Aggregates

Increasing relative humidity

100 μm

Hallet et al., Aerosol Sci. Tech., 1989
What is Black Carbon?

- Byproduct of incomplete combustion
  - fossil fuel burning
  - biomass burning
- Graphitized
- Other names:
  - carbon blacks: produced in controlled conditions
  - soot: atmospheric; contains impurities
  - elemental carbon: measured by thermal analysis
  - black carbon: measured by optical absorption

Internal mixtures at an urban location

Ammonium sulfate particles with soot inclusions

- Lindenberg Aerosol Characterization Experiment (LACE 98)
- 70 km southeast of Berlin
- carbon/sulfate mixtures found in 4-49 volume percent of all particles
- typical soot fraction is 5-10 % by volume; values up to 50% observed

Ebert et al., JGR, 2002
Modeling Global Black Carbon Absorption

15% Tuning factor 2-4

Carbon Emissions Inventory, Uncertainty factor > 2

Dispersion, Transport, and Removal

BC Mass; $M \text{ (g m}^{-3}\text{)}$

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AERONET observations

Park (2003) 15%

Schuster (2005)

Sato (2003)

Tuning factor 2-4
What do we know about global black carbon concentrations and absorption?

- Satellite measurements unavailable, so we rely on models.
- Modeled emission inventories are uncertain by at least a factor of 2.
- Measurements for testing models are inadequate:
  - Not enough surface measurements
  - Vertical distribution available only during field missions
  - More measurements are needed
- AERONET provides radiometric retrievals of column aerosol size distributions and refractive index at 180+ locations.
Principle plane comparisons

Fraction of errors

<table>
<thead>
<tr>
<th>AOT</th>
<th>&lt;5%</th>
<th>&lt;10%</th>
<th>&lt; 20%</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt; 0.4</td>
<td>0.73</td>
<td>0.94</td>
<td>0.99</td>
</tr>
<tr>
<td>0.05-0.4</td>
<td>0.68</td>
<td>0.90</td>
<td>0.98</td>
</tr>
<tr>
<td>&lt; 0.05</td>
<td>0.63</td>
<td>0.83</td>
<td>0.91</td>
</tr>
</tbody>
</table>

Averaged over all angles and all wavelengths
Comparison of calculated and measured almucantar radiances for three ranges of aerosol optical thickness. Fraction of errors less than 5, 10, and 20 percent shown below.

<table>
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<tr>
<td>&gt; 0.4</td>
<td>0.66</td>
<td>0.96</td>
<td>0.99</td>
</tr>
<tr>
<td>0.05-0.4</td>
<td>0.83</td>
<td>0.98</td>
<td>0.99</td>
</tr>
<tr>
<td>&lt; 0.05</td>
<td>0.74</td>
<td>0.82</td>
<td>0.86</td>
</tr>
</tbody>
</table>