Interpreting the Ultraviolet Aerosol Index Observed with the OMI Satellite Instrument to Understand Absorption by Organic Aerosols: Implications for Atmospheric Oxidation and Direct Radiative Effects

Melanie S. Hammer¹, Randall V. Martin¹,², Aaron van Donkelaar¹, Virginie Buchard³,⁴, Omar Torres³, David A. Ridley⁵, Robert J.D. Spurr⁶

1. Department of Physics and Atmospheric Science, Dalhousie University, Canada
2. Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA
3. NASA/Goddard Space Flight Center, Greenbelt, MD, USA
4. GESTAR/Universities Space Research Association, Columbia, MD, USA
5. Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, MA, USA
6. RT Solutions, Inc., 9 Channing Street, Cambridge, MA 02138, USA
UV-absorbing aerosols effect atmospheric photochemistry and Earth’s radiation budget

Aerosol absorption in the ultraviolet:

- Estimated to be the second largest source of radiative forcing after carbon dioxide (Bond et al., 2013; IPCC, 2014; Ramanathan and Carmichael, 2008)
- Decreases photolysis frequencies, leading to decreased concentrations of atmospheric oxidants

Atmospheric chemistry models tend to overestimate OH concentrations compared to observations.

Black carbon is typically treated as sole absorbing carbonaceous aerosol

Brown Carbon (BrC):

- Strong absorption in UV, decreases into visible and near-IR
- Emitted to atmosphere through low-temperature, incomplete combustion of biomass and biofuel
- Found to contribute significantly to absorption by biomass burning aerosol
OMI Ultraviolet Aerosol Index

- Method of detecting absorbing aerosols from satellite measurements [Torres et al., 1998; 2007].
- Found to be sensitive to absorption by brown carbon [Jethva and Torres, 2011]

Simulation

We follow the method of Buchard et al., (2015) for simulating the UVAI using the vector radiative transfer model VLIDORT [Spurr 2006]:

\[
UVAI = -100 \cdot \log_{10} \left( \frac{I_{354}^{RAY+AER}}{I_{354}^{RAY} R_{354}^{*}} \right)
\]

\(UVAI > 0\) : indicates absorbing aerosol

Simulate the UVAI for two cases:
- **Base Case**: primarily scattering organic carbon
- **Case 2**: includes absorbing BrC

Simulations are conducted for the year 2007
Base Case UVAI Simulation Underestimates Biomass Burning Absorption

<table>
<thead>
<tr>
<th>Region</th>
<th>Month</th>
<th>( r )</th>
<th>Mean Bias</th>
</tr>
</thead>
<tbody>
<tr>
<td>West Africa</td>
<td>Jan</td>
<td>0.48</td>
<td>-0.57</td>
</tr>
<tr>
<td>South Asia</td>
<td>April</td>
<td>0.46</td>
<td>-0.32</td>
</tr>
<tr>
<td>Southern Africa</td>
<td>July</td>
<td>0.09</td>
<td>-0.97</td>
</tr>
<tr>
<td>South America</td>
<td>Sept</td>
<td>0.40</td>
<td>-0.50</td>
</tr>
</tbody>
</table>

Negative bias not explained by
- Black carbon concentration
- Biomass burning plume height
- Uncertainty in aerosol optical depth
Brown Carbon Properties Derived from OMI

Properties of BrC: very uncertain

- Varying estimates for absorption by BrC and fraction of total primary organic carbon that is brown (BrC/POC)
- Different observations may reflect different burn conditions [Saleh et al., 2014], chemical loss and evaporation of BrC [Forrister et al., 2015; Zhong and Jang, 2014], or brown SOA.

Treatment of BrC in Case 2 Simulation:

- **Exploit:** TOA radiances used in the OMI UVAI retrievals contain implicit information on BrC from actual burn conditions, on BrC that remains after chemical loss or evaporation, on Br-SOA, and on BrC/POC fraction.
  - **Assume:** spectral dependence of 3 between 300 and 600 nm to represent mean from literature

\[
k = c \cdot \rho \cdot \lambda \cdot \left[35.4 \left(\frac{BrC}{POC}\right)^{-1.25} \cdot \exp(-10.5\lambda)\right]
\]

- **Filled circles:** \(k\) values of BrC derived from seven sensitivity simulations that all achieve the same simulated UVAI
- **Background spectrum:** \(k\) values calculated using equation

\(\rho\): density of organic carbon (g \(\mu m^3\))

(we assume \(\rho = 1.3\) g cm\(^{-3}\))

\(\lambda\): wavelength (\(\mu m\))

\(c\): conversion constant \(\left(\frac{1.0 \times 10^{12}}{4\pi} \mu m^2 g^{-1}\right)\)
Introducing BrC to the Simulation Greatly Reduces the Bias with OMI

### Case 2 Simulation with BrC

<table>
<thead>
<tr>
<th>Region</th>
<th>Month</th>
<th>$r$</th>
<th>Mean Bias</th>
</tr>
</thead>
<tbody>
<tr>
<td>West Africa</td>
<td>Jan</td>
<td>0.68</td>
<td>-0.09</td>
</tr>
<tr>
<td>South Asia</td>
<td>April</td>
<td>0.66</td>
<td>+0.0002</td>
</tr>
<tr>
<td>Southern Africa</td>
<td>July</td>
<td>0.63</td>
<td>-0.22</td>
</tr>
<tr>
<td>South America</td>
<td>Sept</td>
<td>0.57</td>
<td>+0.33</td>
</tr>
</tbody>
</table>

### Base Case

<table>
<thead>
<tr>
<th>Region</th>
<th>Month</th>
<th>$r$</th>
<th>Mean Bias</th>
</tr>
</thead>
<tbody>
<tr>
<td>West Africa</td>
<td>Jan</td>
<td>0.48</td>
<td>-0.57</td>
</tr>
<tr>
<td>South Asia</td>
<td>April</td>
<td>0.46</td>
<td>-0.32</td>
</tr>
<tr>
<td>Southern Africa</td>
<td>July</td>
<td>0.09</td>
<td>-0.97</td>
</tr>
<tr>
<td>South America</td>
<td>Sept</td>
<td>0.40</td>
<td>-0.50</td>
</tr>
</tbody>
</table>
### Spectral Dependence of Absorption for Biomass Burning Aerosol When Including BrC Broadly Consistent with Literature Values

\[
\text{AAOD} = k\lambda^{-\text{AAE}}
\]

- **AAE ~ 1:** spectrally independent absorption (black carbon)
- **AAE > 1:** spectrally dependent absorption

**Biomass burning** exhibits an **AAE ~ 2** over the UV to near-IR wavelength region

### Absorbing Angstrom Exponent (AAE) Values for Biomass Burning Aerosol

<table>
<thead>
<tr>
<th>Wavelength range (nm)</th>
<th>Base Case: assuming non-absorbing organic carbon</th>
<th>Case 2: including brown carbon</th>
<th>Mean AAE literature values for biomass burning aerosol</th>
<th>References for the literature values</th>
</tr>
</thead>
<tbody>
<tr>
<td>350-400</td>
<td>1.2 ± 0.00</td>
<td>2.9 ± 0.06</td>
<td>2.5-3.0</td>
<td>Jethva and Torres, 2011</td>
</tr>
<tr>
<td>350-700</td>
<td>1.1 ± 0.09</td>
<td>2.1 ± 0.04</td>
<td>1.9</td>
<td>Kirchstetter and Thatcher, 2012</td>
</tr>
<tr>
<td>450-550</td>
<td>0.9 ± 0.18</td>
<td>2.3 ± 0.1</td>
<td>1.7 ± 0.07</td>
<td>Corr et al. 2012</td>
</tr>
<tr>
<td>400-700</td>
<td>1.1 ± 0.13</td>
<td>1.9 ± 0.03</td>
<td>1.6 ± 0.06</td>
<td>Schnaier et al. 2005, Russell et al. 2010, Corr et al. 2012, Yang et al. 2009, Clarke et al. 2007 Bergstrom et al. 2007, Yang et al. 2009, Kirchstetter et al. 2004 Russell et al. 2010, Rizzo et al. 2011</td>
</tr>
<tr>
<td>300-1000</td>
<td>0.9 ± 0.10</td>
<td>1.7 ± 0.02</td>
<td>1.5 ± 0.2</td>
<td></td>
</tr>
<tr>
<td>400-1000</td>
<td>0.8 ± 0.10</td>
<td>1.3 ± 0.005</td>
<td>1.4 ± 0.02</td>
<td></td>
</tr>
</tbody>
</table>
Absorption by BrC Decreases Tropospheric OH Concentrations in GEOS-Chem by up to 35% Over Biomass Burning Regions

BrC absorption decreases the $O_3 \rightarrow O(^1D)$ photolysis frequency leading to decreased production of OH

- BrC absorption decreases global mean OH
- Methyl chloroform lifetime to OH increases from 5.62 to 5.68 years
- Closer to estimates from observations: 6.0 (+0.5, -0.4) years (Prinn et al., 2005)
Direct Radiative Effect of BrC Absorption is Cooling at Surface and Warming at TOA

- Use GEOS-Chem coupled with the RRTMG radiative transfer model [Iacono et al., 2008], a configuration known as GC-RT [Heald et al., 2014]
- Calculate the DRE of BrC as the difference in the DRE of organic aerosol when including BrC (Case 2) minus the DRE of organic aerosol when assuming non-absorbing organic carbon (base case)

All-Sky DRE Values for 2007

Prior estimates of global all-sky DRE for BrC range from +0.04 to +0.25 W m$^{-2}$ at TOA and from -1.50 to -0.06 W m$^{-2}$ at the surface (Chung et al., 2012; Feng et al., 2013)
Conclusions

- We interpret OMI observations of the UVAI by developing a simulation of the UVAI using VLIDORT coupled with GEOS-Chem aerosol fields.
- Base case simulation: significantly underestimates the absorption in biomass burning regions.
- Simulation including BrC: much more consistent than the base case at reproducing the OMI UVAI over biomass burning regions.
- Inclusion of absorbing BrC into GEOS-Chem: decreases tropospheric OH by up to 35% over major biomass burning regions.
- Calculate global annual mean all-sky TOA DRE values of +0.05 Wm⁻² and values of -0.06 Wm⁻² at the surface for BrC.

Atmospheric Chemistry and Physics Discussions paper: