Observations of Absorbing Aerosol over Bermuda

STM-2023
Motivation: Absorption Profiles during ACTIVATE-Bermuda

Blue Dates = Tudor Hill Spirals

Measurements = Particle Soot Absorption Photometer (PSAP)

Showing all profiles if altitude > 3.0km

Data from 10-s online merge files
Motivation: Optical Properties for 14 June 2023 Profile

Absorbing Aerosol Layer
Scattering Aloft
Clean Shallow MBL
Absorbing Aerosol: 3 Cases

<table>
<thead>
<tr>
<th></th>
<th>14 June 16:05:15</th>
<th>17 June 16:20:35</th>
<th>18 June 14:09:05</th>
</tr>
</thead>
<tbody>
<tr>
<td>GPS Altitude (km)</td>
<td>4.0 – 8.7</td>
<td>4.0 – 8.0</td>
<td>4.5 – 8.0</td>
</tr>
<tr>
<td>CN_{10nm} (scm^{-3})</td>
<td>3436 (919)</td>
<td>7579 (2242)</td>
<td>4292 (377)</td>
</tr>
<tr>
<td>CN_{10nm-nv} (scm^{-3})</td>
<td>3006 (850)</td>
<td>7029 (2115)</td>
<td>3294 (878)</td>
</tr>
<tr>
<td>CN_{3nm} (scm^{-3})</td>
<td>5675 (1530)</td>
<td>12047 (3862)</td>
<td>7489 (679)</td>
</tr>
<tr>
<td>Abs (532nm, Mm^{-1})</td>
<td>7.2 (2.7)</td>
<td>15.0 (4.6)</td>
<td>8.9 (2.1)</td>
</tr>
<tr>
<td>Scat (550nm, Mm^{-1})</td>
<td>1.0 (0.8)</td>
<td>10.1 (18.5)</td>
<td>1.5 (1.1)</td>
</tr>
<tr>
<td>CO (ppbv)</td>
<td>79.7 (7)</td>
<td>93.4 (10.4)</td>
<td>82.7 (6.3)</td>
</tr>
<tr>
<td>Ozone (ppbv)</td>
<td>52.5 (5.7)</td>
<td>74.9 (12.7)</td>
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Average Value (standard deviation)

- CN_{10nm-nv} is heated to 350°C → BC, dust, or sea-salt
- Size-dependent loss is applied to CN_{10nm-nv}
- High non-volatile fraction suggests that most particles contain a BC ‘core’
  - SSA very low for each case
  - CO is not particularly elevated
Absorbing Aerosol: 3 Cases

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<tr>
<td>CN_{10nm} (scm^{-3})</td>
<td>3.126 (210)</td>
<td>7550 (2242)</td>
<td>1202 (257)</td>
</tr>
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<td>CN_{3nm} (scm^{-3})</td>
<td>5675 (1530)</td>
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<td>SSA (550nm):</td>
<td>0.11</td>
<td>0.38</td>
<td>0.13</td>
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<td>Scattering (550nm, Mm^{-1})</td>
<td>10.1 (18.5)</td>
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Non-vol Fraction: 87% 93% 77%

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- Size-dependent loss is applied to CN_{10nm-nv}
- High non-volatile fraction suggests that most particles contain a BC ‘core’
  - CO is not particularly elevated
Mie Theory: Are Low SSA Values Possible?

- Mie simulations from “MiePlot” for 4 cases with monodisperse particle sizes

- Average SMPS size distributions are shown for each case → 40nm mode
  - Small particle size qualitatively supports the observed low SSA values

** Volume mixing based on Schuster et al. (2005)
With good linear correlation, distributions were simply scaled using non-volatile number at each 0.5km altitude.

- Mie Theory to calculate absorption coefficient for BC and 10%-BC cases

Reasonable agreement for absorption suggests slightly coated BC

Measured scattering is too low and uncorrelated
Mie Theory Closure: PyMieScatt Results

- PyMieScatt Mie Code using the same datapoints
- Reverse Mie calculations (ContourIntersection_SD):
  - $SD_{\text{meas}} + Abs_{\text{meas}} + Scat_{\text{meas}} \rightarrow n + k$

- Results consistent with MiePlot
- $k \sim 0.33 - 0.54$
- Scattering still overpredicted.
Mie Theory Closure: PyMieScat Results

- Calculate Phase function (SF_SD):
  - \( \text{SD}_{\text{meas}} + n_{1.52} + k_{\text{calc}} \rightarrow \text{phase function} \)
- Normalize phase function
- Calculate backscatter:
  - \( b_{180} = \left[ p_{180} \ast \text{Scat}_{\text{calc}} \right] / 4\pi \)
- Calculate Lidar Ratio:
  - \( S = \left[ \text{Scat}_{\text{calc}} + \text{Abs}_{\text{meas}} \right] / b_{180} \)
Mie Theory Closure: PyMieScat Results

- Also ran reverse Mie calculations unconstrained
- 73% of cases did not converge on a valid solution

- k values similar to [n=1.52] case, suggesting insensitivity to real part (n)
- Retrieved n values are low: 1.31-1.48
- Lidar ratios still high
Absorbing Aerosol: Remote Sensing Perspective

Slide 11

Absorbing Aerosol: Remote Sensing Perspective

17-Jun-2022

in-situ: 25 ± 23 Mm⁻¹

14-Jun-2022

in-situ: 8 ± 4 Mm⁻¹
Absorbing Aerosol: Remote Sensing Perspective

**in-situ:** 121-125

**in-situ:** 98-120
All Campaigns: Average Absorption (532nm)

- DC3 (May - June 2012)
- SEAC4RS (Aug - Sept 2013)
- NAAMES-1 (Nov 2015)
- NAAMES-2 (May - June 2016)
- NAAMES-3 (Sept 2017)
- ACTIVATE-1 (Feb - Sept 2020)
- ACTIVATE-2 (Jan - June 2021)
- ACTIVATE-3 (Nov 2021 - May 2022)
- ACTIVATE-Bermuda (June 2022)
All Campaigns: Absorption and CN$_{10nm\text{-}nonvol}$

Number Concentration (scm$^{-3}$)

Absorption (532nm, Dry RH, Mm$^{-1}$)

Abs

CN$_{10nm\text{-}nonvol}$

GPS Altitude (km)

East Longitude (°)

North Latitude (°)

DC3 (May - June 2012)
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NAAMES-1 (Nov 2015)
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ACTIVATE-1 (Feb - Sept 2020)
ACTIVATE-2 (Jan - June 2021)
ACTIVATE-3 (Nov 2021 - May 2022)
ACTIVATE-Bermuda (June 2022)
1. Mie Theory simulations were able to reproduce observations using small, mostly-BC particles
   ** Measurements are self-consistent
     • Why is scattering still too low?
     • Treatment of BC shape (Schlosser)
2. HSRL sees clean conditions for event days, more quantitative analysis is necessary (especially lidar ratio)
3. Absorption events are not obvious in past datasets
   • PSAP measurements may not be sensitive enough

How do we explain the amount and extent of this absorbing aerosol?
   • Biomass burning smoke?
   • Commercial Aircraft Emissions?

FIREX data: LAS = NASA, UHSAS = NOAA
ACCESS data: Moore et al. (2017)